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Hydrocarbons, Polychorinated Diphenyls, and DDE in Mussels and Oysters from the U.S. Coast 1976-1978 - the Mussel Watch

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Monthly temporal changes of factors of two to ten were found for organic pollutants in mussels from the Narragansett Bay station. Concentrations of PCBs and fossil fuel hydrocarbons for some urban stations were one to two orders of magnitude higher than those in remote areas. The northeast "megapolis" of the U.S. coast from the Chesapeake Bay area to Boston, Massachusetts clearly shows elevated concentrations of PCBs and fossil fuel hydrocarbons. The composition of aromatic hydrocarbons in samples with elevated concentrations shows both the influence of oil spill or chronic oil inputs and pyrogenic sources.

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by

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ABSTRACT

Mytilus edulis, *Mytilus californianus*, *Crassostrea virginica* and *Ostrea equestris* were sampled at 90 to 100 stations around the United States coastline during each of three years - 1976, 1977, 1978. Data for concentrations of PCB, DDE, total hydrocarbons, gas chromatographically unresolved complex mixture hydrocarbons, and selected aromatic hydrocarbons are presented for most of the samples. Similar data for monthly samples of *Mytilus edulis* from Narragansett Bay, Rhode Island, U.S.A. and *Mytilus californianus* from Bodega Head, California, U.S.A. and laboratory intercalibrations are presented and discussed.

Monthly temporal changes of factors of two to ten were found for organic pollutants in mussels from the Narragansett Bay station. Concentrations of PCBs and fossil fuel hydrocarbons for some urban stations were one to two orders of magnitude higher than those in remote areas. The northeast "megapolis" of the U.S. coast from the Chesapeake Bay area to Boston, Massachusetts clearly shows elevated concentrations of PCBs and fossil fuel hydrocarbons. The composition of aromatic hydrocarbons in samples with elevated concentrations shows both the influence of oil spill or chronic oil inputs and pyrogenic sources.

INTRODUCTION

The "Mussel Watch" concept of using bivalves - primarily Mytilus species - as sentinel organisms for coastal chemical pollution has been described by several authors (e.g., see N.A.S., 1980; Phillips, 1980). The rationale and initial one year results for the program leading to the results reported here have been set forth by Goldberg et al. (1978). In this report we present and discuss the results of analyses for DDT family compounds, polychlorinated biphenyls and fossil fuel hydrocarbons for 1976-1978 (Years 1,2,3 of "Mussel Watch"). Data on trace metals and transuranics will be presented in other papers.

The analytical methodology for determining low concentrations of organic pollutants in marine organisms has been, and continues to be the subject of continuing intense research (NOAA, 1979; N.A.S. 1980). This was especially the case for fossil fuel hydrocarbons (NAS, 1975; Farrington, 1980). No single method of analysis could be used to obtain data on all possible parameters indicative of fossil fuel contamination. We decided to proceed with the best available methods in each of two laboratories and to continue to develop methods as the project progressed. Intercalibration exercises were used to measure comparability of data obtained in each laboratory.

Analytical instrument capabilities coupled with other research made it possible for one laboratory to make some fossil fuel hydrocarbon measurements not possible in the other laboratory at that time. In addition, unanticipated funding constraints made it impossible to obtain data on one-third of the Year 3 samples. Data for high resolution gas chromatography analyses for these samples will be reported elsewhere. Despite these restrictions, we

think the available data are of importance to considerations of the status of chemical pollution of the U.S. Coast and thus we present some the data and interpretations in this report.

METHODS

Collection: Sites and Procedures

The procedure for collection for the three years has been set forth in Goldberg et al. (1978). Briefly, a team of two collectors traversed the U.S. Coast and collected samples of mussels (Mytilus edulis, and Mytilus californianus) and oysters (Crassostrea virginica, and Ostrea equestris) at the stations listed in Table 1, and shown in Figure 1. A series of monthly samples (Mytilus edulis) were collected in Narragansett Bay, Rhode Island by personnel of the U.S. EPA, Environmental Research Laboratory, Narragansett, Rhode Island and monthly samples of Mytilus californianus were collected at Bodega Head, California by personnel of the Bodega Marine Laboratory, University of California. The serial collections at the Narragansett Bay site were interrupted during the 1976 winter season, by ice conditions and the mussel population at the site did not survive the ice conditions. A new station was established in 1977 and 1978 with a more complete monthly collection sequence. Table 1 provide station information for the Bodega Head and Narragansett Bay sites.

Unfortunately, it was difficult to locate suitable populations of oysters to sample in some areas of the Gulf Coast. There was no doubt such populations existed but they were well guarded secrets as far as exact locations. Our collectors purchased a number of sample collections from local fishermen and the general location of the collections as given by the fishermen are listed

in Table 1 with appropriate notation in the footnotes.

Samples were normally taken from a single site at each location, although on the West Coast some composite collections consisting of up to eleven substations were made at six of the sites; these are so noted in Table 1. The populations sampled were large enough to withstand repeated yearly or monthly sampling; i.e. the sample size was very small, usually less than 1% of the total population. Animals of uniform size were collected and where possible were in the range of 5-8 cm length. Pilings or metal buoys were avoided as collection sites, except in one case in Year 1, in order to avoid influencing the animals' chemical pollutant composition by paints, creosote, or other materials applied to those objects. Sample substrates were rock, sand, or mud environments.

Immediately after collection the animals were placed in a freezer and maintained in a frozen state through shipment to the laboratories for frozen storage (-10 to -20°C) and analysis. Samples were placed in aluminum foil and then shipped in styrofoam-lined containers from four to six sites at a time.

Extraction and Analysis

The procedures used in each laboratory are given separately because the techniques varied in their details, although the basic analytical principles are the same.

UNIVERSITY OF TEXAS, PORT ARANSAS MARINE LABORATORY

Shucking and Extraction: 1976 Samples

The frozen bivalve samples were opened and the tissue plus fluid were removed and stored cold in glass jars with Teflon-lined lids until the complete sample or subsample from a given site was shucked. The cold tissue was

ground to a paste in a Virtis homogenizer and then freeze-dried using a Virtis Model 25 SRC freeze-dryer. Blanks consisting of fired sand were used to check for contamination during freeze-drying.

One part of freeze-dried sample of 7 to 40 g was refluxed overnight (17-18 hours) with ten parts of a 1 N KOH in 85% ethanol digestion-saponification solution. The digestate was extracted twice with 50 ml of hexane and twice with 50 mL of benzene. Saturated NaCl aqueous solution was added to those extractions that formed emulsions. The hexane and benzene extracts were combined, concentrated to near dryness by rotary evaporation and resaponified for two hours with 50 mL of the digestion-saponification solution. This solution was then extracted with two 20 mL portions each of hexane and benzene. The extracts were combined, washed with saturated aqueous NaCl solution concentrated by rotary evaporation and taken to dryness at room temperature under a stream of N_2 and constitutes the non-saponifiable lipids.

Column Chromatography

The non-saponifiable lipids were chromatographed on a solvent packed, two layer column of alumina over silica gel 1:1 by weight. Three fractions were collected: fraction 1 (f1) two column volumes of hexane, fraction 2 (f2) two column volumes of benzene, fraction 3 (f3) two column volumes of methanol. The later fraction was not analyzed in this program. These fractions were concentrated for gas chromatographic and gas chromatographic/mass spectrometry analysis.

Gas Chromatographic Analyses

Each of the f1 and f2 fractions were analyzed on 5% FFAP on 60-80 mesh Gas Chrom Q (acid washed) 0.32 cm x 183 cm columns installed in either a Perkin

Elmer Model 900 or Varian Model 3700 gas chromatograph equipped with flame ionization detectors (F.I.D.). Routine operating conditions were: 30 mL/min. He carrier gas flow, temperature programming from 70° to 270°C at 6°C/min. after a six minute hold at injection at 70°C and a final temperature hold of 24 min. at 270°C. On occasion lower initial temperatures and longer initial or final temperature hold times were used.

Integration of peak areas was accomplished with either an Infotronics Model CRS-204 or Columbia Scientific Industries Model CSI-38 digital integrator. Peaks were identified in the gas chromatograms by retention indices where the retention index of n-heptadecane and n-octadecane are 1700 and 1800 respectively. Peaks eluting between n-alkanes were assigned interpolated retention indices, e.g. pristane = 1670 and phytane = 1780.

Gas Chromatography-Mass Spectrometry

The same type of GC column used for GC analyses was installed in a Varian Aerograph Model 2700 GC (modified) coupled to a Dupont Model 21-491 GC/MS interfaced to a Dupont Model 21-094B MS data system. Operating conditions were: source temperature 200°C; electron accelerating potential of 70 volts; ion accelerating potential of 1400 volts. The mass range of 500 to 40 daltons was scanned. Slits and focusing parameters were adjusted for a maximum of sensitivity at m/e 500 giving an estimated sensitivity of 1.5×10^{-9} g hydrocarbon at MW = 282 in the computer reconstructed chromatograms. Various mass spectral libraries were searched for confirmation of spectra and when needed interpretive techniques were used.

1977 Samples

The same extraction and column chromatography techniques were used with

the following minor modifications. The digestion solution was 4 N KOH in 75% ethanol and was extracted with 4 x 20 mL hexane and 1 x 20 mL benzene. The extracts were combined, washed with water, dried over Na_2SO_4 and concentrated to approximately 5 mL on a rotary evaporator. Then 30 mL of n-heptane was added and the total extract was concentrated to 2 mL. Column chromatography was on a 1:4 by weight alumina over silica column.

Gas chromatographic analyses were accomplished on a 0.25 mm (i.d.) x 11 m SP-1000 glass capillary column installed in a Perkin-Elmer Model 910 gas chromatograph. Operating conditions were 2 mL/min. He carrier gas flow; 5 minutes at 60°C with 2°C/min. programming to 210°C and a 10 minute hold. Selected samples were analyzed on a 0.25 mm (i.d.) x 11 m OV-101 glass capillary column installed in the Perkin-Elmer Model 910 GC. Conditions were similar to the analyses on the SP-1000 column except initial and final temperatures were 70° and 250°C, respectively. Electronic integration of peak areas and retention index assignment was as described for the 1976 samples.

GC/MS analyses were the same as for 1976 samples with the exception that selected samples were also analyzed by glass capillary GC/MS using a 0.25 mm (i.d.) x 10 m OV-101 column.

BODEGA BAY MARINE LABORATORY

Shucking

Each sample from the 1976 collection consisted of fifteen individuals chosen at random from a total sample comprising approximately fifty individual mussels. After each mussel was shucked, the shell length was determined. The tissues of the fifteen pooled mussels were weighed and then placed in a small Servall homogenizer and thoroughly homogenized, after which a 20 g portion was

removed for analysis. Another portion of sample was removed, weighed in a tared aluminum foil pan and dried at 60°C until a constant weight was obtained (usually 2-3 days) for dry weight determinations. The remaining tissue was frozen in a glass jar and archived.

From the samples obtained in 1977, at least forty individuals were shucked into a clean glass jar. On the basis of analyses of individual mussels, a sample size of this magnitude was considered necessary to reduce sampling variance below the expected analytical variance (Risebrough et al., 1980). Tissues were homogenized with a Servall Model 17105 stainless steel Omni-mixer or a stainless steel commerical Waring Blender which was thoroughly cleaned and solvent-rinsed before each sample.

Extraction and Cleanup

1976: the 20 g portion of homogenized tissue was placed in a beaker and dissolved over steam with three times its volume of BFM solution, a mixture of acetic and perchloric acids (Stanley and LeFavoure, 1965). After digestion was complete within 2-4 hours, the solution was transferred to a 125 mL separatory funnel and extracted with three 50 mL volumes of nanograde hexane. The combined organic phases were reduced to approximately 10 mL and transferred to a glass centrifuge tube. One mL of concentrated sulfuric acid was added and the tube was stoppered and shaken for one minute; the tube was then frozen in a dry ice/acetone slurry, and the organic phase was decanted into a graduated cylinder. The tube was then rinsed with three small volumes of hexane which were added to the extract in the graduated cylinder.

For the 1977 samples, the following procedure was used. Approximately 60-100 mL of the homogenate were freeze-dried with a Virtis D10-020 freeze

dryer with an activated charcoal filter between the vacuum pump and the freeze dryer unit. Initial blanks were unacceptable, but placing a plug of polyurethane foam in the glass adaptor reduced contamination to levels considered acceptable over a 24 hour period. The plugs were replaced after 24 hours if freeze-drying were continued for a longer period. The ground glass joints were modified with a groove cut around their circumference which accepts a Viton O ring: the vacuum seal was improved and sticking of the flask to the adaptor was prevented. Homogenates were placed in 1000 mL Erlenmeyer flasks covered internally with a layer of ice to prevent sticking of the dried tissue to the glass. The dry tissue was ground with anhydrous sodium sulfate and Soxhlet-extracted with benzene. After removal of the benzene by rotary evaporation, the lipid was taken up in hexane.

Florisil (Supelco 6/100 mesh) was activated at 250°C for 12 hours, deactivated with 0.5% water, allowed to equilibrate by standing for at least 15 hours, and used within three days. The column (16.5-cm florisil and 1.2-cm sodium sulfate in a 22-mm i.d., 400 mm long chromatographic column) was packed dry. After loading the sample, 150 mL of hexane was applied and the first 105 mL was collected as fraction 1. Three hundred mL of 30% methylene chloride in hexane was then applied and collected as fraction 2. Two hundred mL of 50% methylene chloride in hexane was then applied and eluted as fraction 3. PCBs elute in fraction 1 of this procedure.

Gas Chromatographic Analysis

The sample extracts were analyzed with a Tracor MT-220 gas chromatograph equipped with 2-m, 4-mm i.d., glass columns containing a mixture of 1.5% SP-2250/1.95% SP-2401 on 100/120 mesh Supelcon (AW-DMCS) or 3% OV-1 on 100/120

mesh Supelcoport (Supelco, Inc.) and Ni-63 electron capture detectors. Additionally, sidearm saponification columns (Miller and Wells, 1969) were utilized to convert p,p'-DDD and p,p'-DDT, if present, to their ethylene derivatives, thus providing confirmation of their identify. PCB was quantified by comparing the combined heights of three peaks eluting after p,p'-DDE and with relative retention times of 1.25, 1.48, and 1.74 with the same peaks in an Aroclor 1254 sample.

WOODS HOLE OCEANOGRAPHIC INSTITUTION

Shucking

Twenty-five to one hundred and fifty frozen bivalves from each station were opened in the frozen condition and the tissue plus frozen fluid removed and placed in a clean glass jar. The samples from each station were homogenized in a Tekmar tissumizer. A portion of the homogenate was freeze-dried and the rest was stored frozen at -20°C until analyzed within 1-8 weeks.

Chlorinated Hydrocarbons: Extraction and Analysis

Year 1 and Year 2 - 1976, 1977

The freeze-dried material (∿ 2 g) was placed into a small Virtis homogenizer flask (125 mL capacity) equipped with a nylon impellor along with approximately 12 g of pre-extracted .5-mm glass beads. The chlorinated hydrocarbons were extracted using hexane (1 x 50 mL, 2 x 25 mL) and filtered through pre-extracted glass fiber filter paper, rinsing the filter with 25 ml of hexane. The hexane extract was concentrated on a Buchi rotary evaporator to approximately 1 ml at ∿ 20°C, being careful not to take to complete dryness.

Column chromatography was accomplished on silica gel (100/200 mesh) deactivated 5% with water and topped with alumina (∿ 200 mesh) also deactivated 5%

with water. The glass column was 9-mm i.d. x 35-cm long with a 300 mL reservoir and initially filled with 100 mL of hexane. Then 5 g of silica gel and 5 g alumina were sequentially poured in allowing the air to escape and tapping the column for 1-2 minutes to promote settling. The solvent dripped through until level with the top of the packing.

The extract was loaded on top of the column, rinsing the sample flask four times with 1 mL of hexane, and allowing each rinse to wash through to the top of the packing before adding the next rinse. A final 10 mL of hexane was added and allowed to drip through. All of this hexane was discarded. Finally, 50 ml of 20% toluene, 80% hexane was added to the column and collected. This column extract was concentrated for GC analysis.

Gas Chromatograph Operating Conditions

Column packing - 1.5% OV-17/1.95% QF-1 on Chromosorb W HP 100/120 mesh. Column 2-m length 2 mm i.d. glass. Oven temperature isothermal at 190°C. Injection temperature at 225°C. Detector temperature at 300°C for Ni-63 only. Column flow at 20 mL/min. Auxilliary gas at 40 mL/min. (total of ∿ 60 ml/min.). Carrier gas, pulse mode, 95% arg., 5% methane.

Year 3 - 1978

Samples were extracted using the hydrocarbon extraction procedure given below. Equal portions of the column chromatographic fractions f1 + f2 were combined and analyzed by gas chromatography with electron capture detection. DDT compounds were analyzed as total DDE.

A Perkin Elmer Model 900 gas chroamtograph or Packard Model 407 gas chromatograph, both equipped with a Ni-63 electron capture detector, were used for these analyses. Recoveries of PCBs and DDT family compounds spiked to samples

was 80 to 90% or better. Quantiation of DDT family compounds was by comparison of peak heights for the samples with standards. Quantitation of PCBs was by comparison of five to eight peaks for the samples with the same peaks in chromatograms of Aroclor 1254 and, when applicable, Aroclor 1242.

Hydrocarbons: Extraction and Analysis

Approximately 15 gm of wet tissue homogenate was placed into a 50 mL centrifuge tube with a Teflon lined screw cap. Ten mL 6 N NaOH, prewashed with n-hexane, were added and the contents shaken vigorously for one minute. After one hour the tube was again shaken vigorously for one minute, then placed into an oven at 30°C for 18 hours.

Extraction of the hydrocarbons was accomplished by adding three separate aliquots of ether (1 x 15 mL, 2 x 10 mL) and shaking vigorously for two minutes. Centrifugation was used to clearly separate the phases. The ether extracts were combined and evaporated almost to dryness prior to column chromatography. A few mL of hexane were usually added before evaporation, alleviating the need to go to complete dryness.

Column chromatography was accomplished using a column of dimensions and silica and alumina as described above except for the elution and fraction collections. Fraction 1 was collected by elution with 15 mL of hexane after the extract was placed on the column. Fraction 2 was eluted with 20 mL of 10% toluene and 90% hexane followed by 20 mL of 20% toluene and 80% hexane. These fractions were concentrated by rotary evaporation prior to gas chromatographic analysis.

Gas Chromatography

Both column chromatography fractions (f1 and f2) were analyzed by glass

capillary gas chromatography on a 0.32-mm i.d. 20 m SE-54 or SE-52 column installed in a Hewlett Packard Model 5840 A GC or Model 5710 GC equipped with split/splitless injectors. Operating conditions were splitless injection at 40°C until solvent peak eluted then rapid heating to 70°C and programmed 4°C/min to 240°C. Gas flow rate was 2-3 mL/min helium.

Compounds were quantified by comparison with the peak heights of standards analyzed using the same conditions as sample analyses. The peaks were not measured by automatic integration as the data we have obtained recently with automatic integration of this type of sample has not proved to be reliable. Recoveries were calculated using internal standards n-$C_{14}H_{30}$, n-$C_{22}H_{46}$, n-$C_{28}H_{58}$, hexamethylbenzene and hexaethylbenzene and by measuring recoveries of the BLM hydrocarbon reference mixture added to two portions of subsamples.

Recoveries were: n-$C_{14}H_{30}$, mean = 63.5% ± 15.7 n=126, n-$C_{22}H_{46}$ mean = 93.8% ± 19.0 n=126, n-$C_{28}H_{58}$ mean = 106% ± 23.2 n=126, hexamethylbenzene mean = 32% ± 20 n=85, hexaethylbenzene mean = 56% ± 27 n=49. The n-$C_{14}H_{30}$ recoveries are somewhat low and were expected due to the volatilization of lower molecular weight hydrocarbons up to n-$C_{14}H_{30}$ during the solvent evaporation and concentration steps. The same reasoning applies for hexamethylbenzene. The recovery for n-$C_{28}H_{58}$ averaged above 100% due to occasional samples where naturally occurring n-$C_{28}H_{58}$ or a hydrocarbon with similar retention index co-eluted with the peak. The recoveries presented for n-$C_{14}H_{30}$, n-$C_{22}H_{46}$, and n-$C_{28}H_{58}$ are for spiking during extraction and carrying through the entire analyses including gas chromatography. The recoveries for hexamethylbenzene and hexaethylbenzene

are for the entire extraction and analysis procedure including GC/MS. A comparison of recovery values for hexamethylbenzene and hexaethylbenzene for the procedure through gas chromatography with recoveries including the additional step of GC/MS show an additional ten to twenty percent loss is associated with the manipulations of going to GC/MS. This is probably due to the small amounts of compounds we are handling and losses to glassware during concentration and transfer steps.

We have noted recoveries have been better for both hexamethyl- and hexaethylbenzene spiked to Mytilus edulis contaminated with No. 2 fuel oil at 60 to 200 μg/g dry weight concentrations. This implies that a "carrier effect" is operative. The greater the amount of hydrocarbons present, the higher the recovery of internal standards due to smaller losses to glassware already "deactivated or coated" with some of the other compounds. We have reported our recoveries for other spiking experiments including a mixture of alkanes and aromatic hydrocarbons in the n-$C_{14}H_{30}$ to n-$C_{28}H_{58}$ and naphthalene to pyrene range elsewhere (Farrington et al., 1982). Recoveries for selected aromatic hydrocarbons were: methylnaphthalene = 63%, C_2-naphthalene = 62-74%, phenanthrene = 100%, pyrene = 107%.

We are reinvestigating the various extraction and isolation procedures with the intent of improving precision, accuracy, and recovery of compounds. The data presented in the tables in this report are not corrected for recovery.

The unresolved complex mixture signal was measured by planimetry.

Identification of peaks was based on retention indices of compounds and coinjection, and for aromatic hydrocarbons additional identification by mass spectra.

Quantitative Gas Chromatography-Mass Spectrometry

The aromatic/alkene fraction (f_2) were analyzed on a Finnigan 1015 quadrupole mass spectrometer coupled to a Varian gas chromatograph equipped with a split/splitless injector. A 17 m x 0.32 mm i.d. SE-54 glass capillary column was installed in the system and operated as follows: Injection was at 40°C with 30 seconds in splitless mode, then rapidly brought to 70°C and programmed to 240°C at 4°/minute. Helium carrier gas was at 8 psig. The mass spectrometer operating conditions were: electron energy, 40 eV; ionization current, 500 μA; mass scans, 40-500 daltons in three seconds. Quantitative analyses were achieved by analyzing known standard solutions of aromatics using the same conditions and plotting a response curve of amount vs. ion current. Ion currents were integrated from mass plot data using an integration program in the Riber 400 software package for the Finnigan 1015 data system.

The aromatic hydrocarbons measured were naphthalene, methylnaphthalenes, dimethyl- and ethyl- (C_2-) naphthalenes, phenanthrene, methylphenanthrenes, dimethyl- and ethyl- (C_2-) phenanthrenes, fluoranthene, pyrene, dibenzothiophene, and methyldibenzothiophenes. The phenanthrenes were tentatively separated from corresponding anthracenes by glass capillary GC retention time. These compounds were chosen among the thousands of aromatics present in fossil fuels because (i) they are often major constituents in fossil fuels, (ii) many of them are found in fossil fuel combustion products, (iii) standards were available for instrument calibration, and (iv) the relative ratios of some of the compounds could provide insight into sources of hydrocarbons in the bivalves and their habitat.

RESULTS AND DISCUSSION

Data Set

The entire data set is too large to include here and thus we have used representative groups of data to illustrate a given interpretation or in some more important cases the entire subset of a given type of data. Table 2 gives a synopsis of the data available. A more detailed data file report is in preparation.

Intercomparison and Precision

The results of the DDE and PCB analyses for the serial samples taken at Bodega Head, California and Narragansett, Rhode Island are presented in Tables 3 and 4. In one laboratory we checked on the consistency of agreement of duplicate analyses throughout the first three years of analyses in order to gage reproducibility of analytical techniques. The agreements between duplicate analyses were (within 20% of the average) quite good except for three sets of PCBs in the early set of analyses for the Bodega Head mussels as indicated in Table 3. Since the duplicates for DDE in the same samples agreed there is no explanation we can think of other than inhomogeneity in the subsamples which results in a discrepancy for PCBs, but not for DDE. Blanks run along with these samples showed very low PCB contamination - much lower than could account for the discrepancies.

There was also good agreement (within 20% of the average) between laboratories for PCB concentrations in the large mussel homogenate from Narragansett Bay used as an intercalibration sample (Table 4). Thus, we have reasonably good intercalbration for measurements of PCB concentration one order of magnitude apart and within the range of concentrations we have

usually found in mussels and oysters.

The results of intercalibration measurements between the two laboratories analyzing hydrocarbons are presented in Tables 5 and 6. The measurement of the f1 unresolved complex mixture and quantitative measurement of several polynuclear aromatic hydrocarbons were not attempted in one of the laboratories except for a very few samples. Thus, we do not have intercalibration data for these nor do we compare these data for about half the Gulf Coast and most of the West Coast samples. Examples of the agreement of duplicate hydrocarbon analyses of subsamples of homogenates of mussels and oysters are presented in Tables 5-8. In all cases except a few of the f_1 alkanes (Table 5), the agreements are good considering the state of the art of this type of analysis (Wise et al., 1980). The precision of the measurement of the aromatic hydrocarbons by quantitative GC/MS could probably be improved by using selected ion monitoring for molecular ions of the compounds to be measured. This would allow an increase in the number of scans per peak over the fuller range scans of 40-400 or 500 daltons we employed here. However, valuable mass spectral information needed to confirm the identity of the compound measured would be lost. Despite the high resolving power of the glass capillary columns we experienced a number of occasions when fragmentations from olefinic hydrocarbons of biogenic origin would have interfered with and given false measurements for PAHs. The ability to check the spectra allowed us to eliminate these spurious peaks from integration of PAH ion currents. Repeat analyses of the samples to obtain both increased selected ion monitoring precision and full scan range spectra was prohibitive in cost at the time of these measurements, given the exploratory nature of

these measurements in this program. This is not the case now and better precision will be achieved in the future.

The agreement between the two laboratories measuring hydrocarbons for n-alkanes and branched alkanes in a variety of samples over a wide range of concentrations and a span of several years (Tables 4 and 5 in Farrington et al., 1974) is not as good as for DDE and PCB measurements. Several problems of precise measurements of small individual n-alkane gas chromatography peaks in the presence of an unresolved complex mixture signal have been encountered, e.g. higher molecular weight C_{22} and C_{30} n-alkanes (Tables 5 and 6). When peaks are clearly resolved and substantially elevated above the unresolved complex mixture signal then agreement is improved, e.g. pristane (Tables 4 and 5 in Farrington et al., 1974).

Our laboratories have also participated in a U.S. National Bureau of Standards sponsored working intercalibration where our data agree reasonably well with several other laboratories (Wise et al., 1980), although in that case agreement was generally no better between all laboratories than we have achieved between two laboratories. Obviously, the state of the art needs improvement. Furthermore, there can be improvement in the DDE and PCB measurements and there is a need for further intercalibration for a variety of concentrations for many more organic pollutants in several different tissue matrices. Data from a more extensive intercomparison exercise within the U.S. Mussel Watch program will soon be available.

Monthly Concentration Changes

The data for PCBs and DDE concentrations in Bodega Head and Narragnsett Bay mussels are given in Tables 3 and 4. There is variability in concentration

data of factors of two to eight for the Bodega Head samples and two to four for the Narragansett Bay samples for any given year's data. n-Alkanes such as n-heptadecane (n-$C_{17}H_{36}$) and branched alkanes such as pristane vary in concentration by an order of magnitude and sometimes more in comparing monthly data for Bodega Head and Narragansett Bay (Tables 9 and 10). The total resolved alkanes and alkenes in fraction 1 from column chromatography as quantified by GC F.I.D. varies by as much as an order of magnitude. This is not surprising given the fact that many of these compounds are biogenic in origin either in the mussels or in their habitat (Farrington et al., 1976) and their concentrations would be influenced by fluctuations in biological processes.

Only traces or no discernible fl unresolved complex mixture (UCM) of alkanes and cycloalkanes were found in the Bodega Head samples. The UCM in the Narragansett Bay mussels varied in concentration by a factor of two to four within a given year (Table 10). The composition of the alkane and cycloalkanes in the mussels fluctuated markedly for these samples as illustrated by the fraction 1 and fraction 2 gas chromatograms of the 1976 data set given in Figures 2 and 3. Similar fluctuations in resolved GC peaks were observed for the Bodega Head samples.

Aromatic hydrocarbon concentrations varied by factors up to an order of magnitude within a given year for the Narragansett Bay site (Table 11).

U.S. Coast Data

Halogenated Hydrocarbons

The concentrations of PCBs and DDE in mussels and oysters from the U.S. East, Gulf, and West Coast samples for 1976 and 1977 are given in Tables 12

and 13. Results available for three years data, 1976-1978, at the same stations are given in Table 14. For some samples the PCB concentrations were high relative to the DDT family compounds (DDT, DDD, DDE) such that PCB peaks in the gas chromatograms interfered with DDE measurements given the analytical separations employed for the 1976 and 1977 samples.

The known "hot spot" of DDT residues of the Southern California Coast (Risebrough et al., 1980) is apparent in the 1976 and 1977 data (Table 12). The elevated DDE concentrations in the Los Angeles area can be traced to the Whites Point sewer outfall discharge of wastewater which included DDT manufacturing wastes (Berkhiser, 1974; Risebrough et al., 1980). DDE concentrations in mussels and oysters from the remainder of the West Coast and the East and Gulf Coast stations were low - (2 to 40 x 10^{-9} g/g dry weight; Tables 12-13) and probably reflect the remnant DDT residues from agricultural use in the years prior to 1976. There is fairly good agreement of the 1976 and 1977 data sets (Tables 12 and 13) if fluctuations recorded for the variability in DDE and PCB concentrations in monthly Narragnsett Bay and Bodega Head samples are taken into account (Tables 3 and 4).

There are some stations which had relatively low concentrations of PCBs in mussels and oysters at the 3 to 50 x 10^{-9} g/g dry weight range (Tables 12 and 13). These stations are located in fairly remote, relatively pristine locations and we think the concentrations represent a range of background values from the widespread use and general dispersal of PCBs due to mobilization after release to the environment and transport via aeolian, fluvial and coastal oceanic processes. Stations with high concentrations of PCBs in the greater than 200 x 10^{-9} g/g dry weight range were located in or near

industrialized areas of the coast associated with large cities, e.g. San Diego, San Pedro Harbor (Los Angeles), San Francisco, Boston, New York. There were numerous stations with PCB concentrations less than 200 but greater than 50 x 10^{-9} g/g dry weight.

The PCB concentration data for mussels and oysters strongly indicate elevated regional values for the Northeast U.S. Coast from Chesapeake Bay to Boston when comparing data for the same species (Mytilus edulis) north of Boston and in Oregon and (Crassostrea virginica) from the Southeast and Gulf Coasts. A similar pattern of regionally elevated PCB concentrations was noted for the Southern California Coast from San Diego to Point Fermin north of Los Angeles. The concentration of PCBs in the 300 x 10^{-9} g/g dry weight category for oysters at Biloxi, Mississippi is probably associated with its proximity to an industrial canal (Farrington, personal observation), although this bears further investigation. Unfortunately, due to logistical constraints several of the Gulf Coast station samples in the vicinity of the Mississippi River delta have not yet been analyzed for PCBs.

The PCB concentrations for the 1976 and 1977 samples from the same location seem to agree fairly well (Tables 12 and 13). This is illustrated by plotting the 1976 vs. 1977 data (Figure 4). The correlation of linear regression is quite good if the 1977 composite vs. the 1976 individual samples at San Diego Harbor are excluded from the calculation.

A three year data set of PCB concentrations is available for fourteen stations (Table 14). Again there appears to be only minor, if any, concentration changes from 1976 to 1978 if the monthly fluctuations noted for Bodega Head and Narragansett Bay are taken into account (Tables 3 and 4). The lower con-

centrations of PCBs for the Beaufort, Charleston, and Pass Christian locations in 1978 may be the beginning of a downward trend again but more time series data is needed.

We had reason to suspect elevated PCB concentrations might be present in mussels from New Bedford Harbor, Massachusetts as there were two electrical components manufacturing plants which had operated in the area for sometime and presumably had released some PCBs to the environment. DeLappe et al. (1980) had obtained water samples in the area and others (see explanation in Farrington et al., in prep.) had sampled the sediments in the area at selected locations. High PCB concentrations were reported for both sediment and water. Because of this, a station was added to the U.S. EPA Mussel Watch collection in this area in 1978. PCB concentrations for mussels from New Bedford Harbor and the nearby region (Figure 5) are given in Table 15. Clearly there is a severe PCB pollution problem in the New Bedford Harbor area. This is the only station where strong indication of PCB mixtures other than the 1254 type was given by packed column gas chromatography. Concentrations of Aroclor 1242 or 1016 mixture contributions equal to Aroclor 1254 contributions are present in the harbor mussels. This is probably due to the close proximity to the source of input of PCBs. Increasing distance from the source would favor the loss of the lesser chlorinated isomers due to their greater ease of biological and chemical degradation and/or loss to the atmosphere (EPA, 1976; NAS, 1979). All other stations at which we have measured PCBs in bivalves show only the 1254 type mixtures by packed column estimates. As we will discuss later, we have switched to glass capillary gas chromatography to provide a more quantitative measurement of the absolute and relative concentrations of PCB isomers.

Based on this Mussel Watch data we have been conducting an investigation into the PCB biogeochemistry in the New Bedford Harbor/Buzzards Bay area (Farrington et al., in preparation).

Hydrocarbons

The total hexane fraction gas chromatogram peak concentrations are given in Table 16 for the 1976 and 1977 samples for most of the Louisiana and Texas Gulf Coast and the U.S. West Coast. This total includes major n-alkanes, branched alkanes, alkenes, cycloalkanes, cycloalkenes isolated from column chromatography by the methods (see Methods section) used by the Port Aransas Marine Laboratory, University of Texas. Similar data for the U.S. East Coast and the remainder of the Gulf Coast samples are given in Table 13. There are severe limitations on the interpretation of this measurement because hydrocarbons of biogenic origin can and often do, contribute substantially to this total. These contributions depend on the rates of formation and destruction of biogenic hydrocarbons in the mussels, oysters and their habitat. Problems associated with using this as a parameter for fossil fuel hydrocarbon pollution detection have been thoroughly discussed by Farrington et al. (1976, 1977, 1980), among others, and we will not go into details here. Unfortunately, for the sample locations listed in Table 16 we could not obtain integrated measurements of the total gas chromatograms unresolved complex mixture (UCM) in 1976 and 1977. However, we can provide an indication of the type of contribution of the UCM to the total chromatogram. The use of the UCM as an indicator of the presence of fossil fuel hydrocarbons is discussed by Farrington et al. (1976, 1977, 1980), Wakeham and Farrington (1980), among others. Data for this parameter for East Coast and the remainder of the Gulf Coast samples in

1976 and 1977 are given in Table 13. The limited data available for 1978 samples analyzed to date are given in Table 17. Illustrations of gas chromatograms with and without measurable UCM signal are given in Figure 6. Concentrations of UCM hydrocarbons have been measured for ten selected stations for the 1976, 1977 and 1978 sample collections and this data is presented in Table 17. The year to year agreement is good given the monthly fluctuations seen for the Narragansett Bay station. No discernible time trends of either increasing or decreasing concentrations are evident.

Concentrations of several aromatic hydrocarbons and dibenzothiophenes were measured by quantitative GC/MS for the 1976 and selected 1977-78 East Coast and several of the Gulf Coast samples (Table 18). As stated previously, unanticipated problems with the GC/MS in one laboratory prevented more extensive quantitative aromatic hydrocarbon analyses. The total concentrations of aromatic hydrocarbon concentrations are given in Tables 13 and 18. Quantitative GC/MS measurement of aromatic hydrocarbons for 1976, 1977 and 1978 are available for only three stations in addition to Narragansett Bay and these are given in Table 19. The agreement between 1976, 1977 and 1978 measurements was good for the Rockaway Point and Beaufort samples. There was a general decline in concentrations of aromatic hydrocarbons for the Charlestown samples from 1976 to 1978. This requires further investigation to substantiate long-term trends and causes.

The hydrocarbon data set for UCM concentrations, total aromatic hydrocarbons and individual aromatic hydrocarbons indicates several stations in remote areas have non-detectable UCM concentrations and low aromatic hydrocarbon concentrations (Tables 13 and 16). By analogy to the PCB concentrations, we think

the low concentrations of aromatic hydrocarbons represent the range of background values due to widespread use and dispersal of fossil fuel compounds and some contribution from natural sources such as oil seeps along sections of the California coasts and forest or grass fires as is the case for aquatic sediments in remote locations (see reviews by Farrington, 1980; Neff, 1979; LaFlamme and Hites, 1978, 1980; Wakeham and Farrington, 1980).

Total UCM hydrocarbon concentrations were factors of 30 to 50 higher for samples taken near urban or industrial areas. Totals of aromatic hydrocarbons were factors of 5 to 40 higher for samples near urban and/or industrial areas. Individual or groups of aromatic hydrocarbons had ranges of values similar to totals of aromatics when comparing remote, fairly pristine, areas with urban or industrial locations such as Boston, New York, San Pedro Harbor, San Diego Harbor, San Francisco Bay, Biloxi, Charleston, Narragansett Bay, Galveston (Tables 11, 13, 18-19). There are also indications of regionally elevated concentrations of UCM and aromatic hydrocarbons for the Northeast U.S. Coast (Table 13) from the Chesapeake Bay area to Boston when compared to remote areas in Maine, Washington and Oregon, and Southeastern U.S. However, this regional trend is not as clear as for the PCB data previously discussed.

Examination of the gas chromatograms of the samples from the Pacific and Texas-Louisiana Gulf Coast in 1976-77 revealed that a substantial number contained UCM suspected to be of petroleum origin. In four of the samples fossil fuel hydrocarbon pollution was confirmed by qualitative and GC/MS analyses of aromatic hydrocarbon fractions. These four were oysters from Espiritu Santo Bay, Texas (ESP 770317), oysters from Matagordo Bay, Texas (MAT 770316), oysters from Galveston Bay, Texas (GAL 770214), mussels from Boundary Bay,

Washington (BB 760818). There were also indications of regional fossil fuel contamination in the gas chromatograms of oysters from the Louisiana, Texas Gulf Coast in the 1977 sample set (Table 16) as evidenced by the presence of a very significant unresolved complex mixture signal in the gas chromatograms. Total resolved hydrocarbon concentrations (Tables 13 and 16) do not correlate with other better indicators of fossil fuel hydrocarbons such as the UCM and aromatic hydrocarbon concentrations (Tables 13 and 16) due to the influence of biogenic hydrocarbons.

The absolute and relative concentrations of individual and groups of aromatic hydrocarbons are plotted as bar graphs in Figure 7 for representative samples of oyster and mussels with low and high concentrations of aromatic hydrocarbons. Two important general features of the aromatic hydrocarbon data are illustrated. First, at low concentrations of total aromatic hydrocarbons, phenanthrene often has a higher concentration than C_1- and C_2-phenanthrenes. Fluoranthene and pyrene are usually equal to or higher in concentration than phenanthrene. In general, the qualitative composition is similar to that reported for these compounds in marine sediments from remote and/or pristine locations and sediments near industrial areas where there seems to be predominant urban pyrogenic sources for aromatic hydrocarbons in sediments (Wakeham and Farrington, 1980; LaFlamme and Hites, 1978; Youngblood and Blumer, 1975). However, the qualitative composition of the aromatics in bivalves with the low to medium concentrations of aromatic hydrocarbons is not always a good match with that expected for the pyrogenic source. There is an indication in many cases of a composition which is best explained by a mixture of a pyrogenic source and a petroleum source with dominance of C_2- and C_3-

naphthalenes and C_1- and C_2-phenanthrenes relative to phenanthrene, fluoranthene and pyrene.

There are three possible pyrogenic sources. One is a combined aeolian/fluvial transport of pyrogenic aromatics from urban areas or forest and grass fires (McMahon and Tsoukalas, 1978) to the sampling locations. The second is creosote from wood pilings. Creosote is a pyrogenic product and contains aromatic hydrocarbons with a composition similar to that found in combustion sources (Lake et al., 1979). While only the samples from North San Francisco Bay were collected from creosote wood pilings, numerous pilings of this type have been in place in most U.S. coastal waters for many years. Eaton and Zitko (1979) demonstrated that the influence of the creosote pilings on aromatic hydrocarbons in bivalves decreased with increasing distance from pilings and the effective radius of a discernible effect was only a few hundred meters in the coastal areas investigated. However, it is possible that creosote is the source of some of the low concentrations of aromatic hydrocarbons in the samples from this study.

A third possible source is exhaust from numerous pleasure craft with outboard and inboard engines in coastal areas. This source cannot be substantiated by any measurements we are aware of but can be argued on a theoretical basis. The combustion products should have an aromatic hydrocarbon mix with some similarity to exhaust measured to date from other combustion processes including automobiles (Wakeham et al., 1980). If these are combined with aromatic hydrocarbons from uncombusted fuel in the exhaust, then a mix of the observed proportions of aromatics in the bivalves might occur. Clark et al. (1975) have shown that alkanes from outboard motor exhausts are taken up by

Mytilus edulis and Ostrea lurida in laboratory experiments.

Other sources of petroleum hydrocarbons which could contribute to low concentrations observed in the bivalves - depending on sampling locations - are chronic releases from industrial and municipal effluents, small oil spills, effluents from exploration and production oil rigs, ballast water discharges, natural oil seeps. The acutal relationship of specific sources to samples cannot be discerned because of the complex dynamics of mixing processes in the coastal areas. It suffices for the purposes of this study to show that we have established the existence of low concentration of aromatic hydrocarbons in relatively remote areas of the coast removed from large urban/industrial locations and that plausible sources for these compounds can be suggested.

The second important general feature of the aromatic hydrocarbon data illustrated by the bar graphs in Figure 7 is the predominance of C-1 and C-2 phenanthrenes relative to phenanthrene and often relative to fluoranthene and pyrene in mussels and oysters with the higher concentrations of aromatic hydrocarbons. The predominant source of aromatic hydrocarbons seems to be petroleum, although higher concentrations of phenanthrene, fluoranthene and pyrene could also be partially caused by increased inputs of pyrogenic aromatics relative to the more remote areas. There is data available for aromatics in sediments in several of the general sampling areas in the Northeast U.S. e.g. Boston, Narragansett Bay and the New York Bight in: LaFlamme and Hites (1978); Wakeham and Farrington (1980); Lake et al. (1979); Farrington et al. (1980). The general pattern of aromatic hydrocarbon distributions in the sediments shows a characteristic signature of pyrogenic sources with minor contributions from petroleum - the opposite to that seen in the bivalves sampled during the

same general time period.

We offer the following hypothesis for an explanation of these results for mussels and sediments. The aromatic hydrocarbons in sediments are contributed by several sources - chronic pyrogenic and petroleum releases including small spills. The pyrogenic inputs are primarily associated with particulates formed during or shortly after combustion and release to the atmosphere. Aromatics, and presumably many other compounds associated with them, are strongly associated with or entrapped in these particulates. These tightly bound molecules are not easily removed and taken up by bivalves after ingestion of the particles by filter feeding. Chemical extraction, on the other hand, is a harsher treatment and more readily releases these compounds from particulates in the sediments. Petroleum inputs from chronic releases in effluents or by small spills are accompanied by solubilization in the water and adsorption on detritus or incorportion into phytoplankton (NAS, 1975; Farrington, 1975; Neff, 1979; Connell and Gilbert, 1980-1981). Thus, they are more readily available for uptake by bivalves via particle ingestion or across membrane surfaces.

We may have to modify interpretations of analyses of sediment cores for the historical record of hydrocarbon inputs to certain coastal areas. If petroleum inputs are more readily available for uptake by mussels and oysters relative to pyrogenic inputs then they also might be more readily available for metabolism by marine animals and bacteria in the benthos. Thus, the petroleum inputs might not survive initial deposition and burial as easily as more firmly bound pyrogenic inputs found in aeolian or soil particulates entering estuarine and coastal ecosystems. This interpretation is amenable to testing and we are in the process of initiating such studies.

Interpretations of the petroleum and pyrogenic hydrocarbon data in terms of "hot spots" and time trends have to be approached with caution. We have completed an investigation of a small oil spill at one of the "Mussel Watch" stations during the course of this study (Farrington et al., 1982). The duration of exposure to No. 2 fuel oil was about one day. The biological half-lives for release of the fuel oil alkanes and aromatic hydrocarbons were on the order of 1 to 3 days during the first twenty-one days. This study agrees with other laboratory or acute field exposure data for mussels and oysters recently re- viewed by Marchand and Cabane (1980). Thus, small oil spills or intermittent inputs from effluents with a long time lag between discharges probably results in periods of high and low concentrations. This may explain all or some of the temporal changes in the Narragansett Bay data (Tables 10 and 11) where concentration changes of factors of two to ten were observed.

However, there is good evidence to show that bivalves exposed to long term chronic inputs have very slow release times or biological half-lives once transferred to cleaner environments, suggesting that the mussels and oysters can function as sentinel organisms for areas receiving chronic inputs of fossil fuel compounds (DiSalvo et al., 1975; Dunn and Stich, 1976; Boehm and Quinn, 1977; Farrington et al., 1980, 1982; Marchand and Cabane, 1980).

General Discussion

DDT and PCBs

There is no other U.S. Coast-wide data set for chlorinated hydrocarbons and/or hydrocarbons in mussels and oysters to which we can compare our data with the exception of the chlorinated pesticide data of Butler (1973) for 1965-1972. There are data for U.S. regions and individual estuaries - es-

pecially for pesticides and PCBs. Comparisons are constrained, particularly for hydrocarbons, by the lack of extensive intercalibration between laboratories partly due to a lack of standard reference material. However, with that important caveat in mind it is worthwhile to discuss comparison of some of the available sets of data.

Butler (1973) reported on measurements of DDT family compounds in 8,095 shellfish samples mostly Crassostrea virginica, Crassostrea gigas, and Mercenaria mercenaria and a few samples of Mytilus edulis and Mytilus californianus from estuarine and coastal areas of the U.S. between 1965-1972. Several other investigators reported DDT or DDT metabolite data, e.g. DDE for regional or local coastal areas for the same time period (1965-1972) and extending up to 1975. These reports have been compiled together in a data bibliography for the U.S. Mussel Watch Program (Kidder, 1977). The comparison of our data to Butler's data for DDT compounds yields the same interpretation as a comparison of our data set with each of these other data sets.

Butler (1973) concluded that "DDT pollution in many estuaries, as judged by the magnitude of the residues (DDT family compounds) peaked in 1968 and has been declining markedly since 1970". The percent of eastern oyster samples collected at 10 stations of North Carolina that contained more than 10×10^{-9} g/g wet weight of DDT decreased from over 90% in 1967 to 25% in 1971 (Figure 2 in Butler, 1973). Our data for DDE (Tables 12 and 13) when compared with Butler's data shows that in the time between 1972 and 1976 DDT family compound residues did not increase. If we exclude the special case of the Southern California coastal samples, as discussed previously, there are no stations in our data set where DDE concentrations exceed 100×10^{-9} g/g dry weight

tissue, which is approximately 13 x 10^{-9} g/g wet weight based on our moisture determinations for the samples in our collection. Butler's 1965-1970 data set had 39% of samples with concentrations less than 10 x 10^{-9} g/g wet weight tissue and his 1971 data set had 59% of the samples with less than 10 x 10^{-9} g/g wet weight. Our data show that in 1976 and 1977, 84% and 90% respectively, of our samples contained less than 50 x 10^{-9} g DDE/g dry weight tissue (6 x 10^{-9} g DDE/g wet weight tissue). Butler interpreted his data as indicating that in some areas DDT pollution had become more widespread but this resulted in lower concentrations of residues in the estuarine food web. We conclude from our data and comparison with Butler's data and data of others as compiled in Kidder (1977) that DDE has remained a widespread environmental pollutant. Concentrations of DDE in bivalves appear to have declined in U.S. coastal areas between 1971 and 1976. The most likely and most obvious explanation for the decline is the restriction of the use of DDT accompanied by "dilution" of DDT concentrations by mixing into more of the environment and degradation by biological processes.

Compelling evidence for the ability of bivalves such as oysters and mussels to reflect decreases in environmental levels of DDE has been presented by the investigators of the Southern California Coastal Water Research Project (Berkhiser, 1974) and the California Mussel Watch Program (Risebrough et al., 1980). When the discharge of DDT manufacturing wastes into the Los Angeles County Sanitation District Joint Water Pollution Control Plant was decreased in 1971 the DDE concentrations in Mytilus californianus sampled at Royal Palms State Beach decreased a factor of 64 from 16 x 10^{-6} g/g dry weight in 1971 to 250 x 10^{-9} g/g dry weight in 1978. Concommitant decreases of factors of

5 to 10 for DDE concentrations in mussels were noted for many other California coastal stations (Risebrough et al., 1980).

We expect that the concentrations of DDT family compounds in bivalves along the U.S. coast will continue to decrease over the long term of decades providing there are no new inputs to the U.S. coastal areas as a result of resumption of use of DDT. The lack of discernible differences between the 1976 and 1977 data sets is expected based on the exponential type decrease in concentrations observed after abrupt cessation of inputs to the California coastal area (Risebrough et al., 1980).

The Mussel Watch U.S. coastal PCB data set is unique and no data exists that is comparable to Butler's (1973) DDT data set. A few local and regional studies which have been compiled in the data bibliography (Kidder, 1977) are available but no extensive comparison can be made with DDT compounds. In general, wherever comparisons of local data can be made there is no compelling evidence of decreases in PCB concentrations from 1972 to 1976. Data are not available prior to that time. The one exception is in the Southern California area where PCB data were collected along with the DDT data previously discussed (Risebrough et al., 1980; Berkhiser, 1974). Decreases in PCB concentrations of factors of up to an order of magnitude have been noted for mussels from some locations in comparing 1971 and 1978 data (Risebrough et al., 1980). However, other locations show little or no decrease in concentrations. Most of the observed decreases appear to have taken place between 1971 and 1976. We have noted no significant decreases in PCB concentrations between 1976 and 1977 or 1978 for samples in our data set (Tables 12, 13, 14). Thus, we see no evidence of a widespread decrease in PCB concentrations in U.S. coastal waters as a re-

sult of recent restrictions on use of PCBs. There may have been such a decrease after the ban on open use in 1972 but no data except that from the California data which spans that time period. It is difficult to discern whether the decrease noted for California is peculiar to that area due to changes in sewer outfall operations or is indicative of a more general decrease throughout the U.S. coast.

The DDT data previously discussed suggests that once discharge of persistent chemicals to the coastal and estuarine environment ceases there will be a period of a few years of rapid decrease of concentrations in shellfish followed by a longer period of much slower decrease in contaminant concentrations. Continuing measurements at Mussel Watch stations can document if PCBs will behave in a similar manner. There are still substantial quantities in use or buried in landfills throughout the U.S (EPA, 1976; NAS, 1979). Thus, despite cessation of manufacturing and open use in the U.S., PCB release to coastal and estuarine areas will continue for some time, presumably at a reduced rate compared to release when PCBs were extensively used in open systems.

Comparison of U.S. Mussel Watch data with data for DDT compounds and PCBs in mussels and oysters from European coastal areas as summarized by Holden (1973), Marchand and Cabane (1980), Marchand et al. (1974, 1976) and Cowan (1981) show similar trends of high concentrations of PCBs near industrial areas and lower concentrations in more remote areas for the time period 1968 to 1974. The highest concentrations reported were along the northwest Mediterranean coast in the range of 1 to 10 x 10^{-6} g/g dry weight for Mytilus galloprovincialis collected in metropolitan areas. Generally, these 1973-1975 concentration levels are higher by one order of magnitude than the 1976-1978 data for

the U.S. coast, although the values for New Bedford, Massachusetts, USA (Table 15) are still the highest PCB concentrations (32×10^{-6} g/g dry weight) found to date.

We caution again that comparison of results from several laboratories as we have discussed above must be tempered by the realization that inherent differences in analytical procedures between all laboratories concerned may be the cause of some of the observed variability. In addition, a variety of environmental, biological, or fluctuating input factors may account for some of the differences observed (see review by Phillips, 1980) and data for Narragansett Bay (Table 4).

Petroleum, Fossil Fuel Compounds and Biogenic Hydrocarbons

Connell and Gilbert (1980-81), Marchand and Cabane (1980), Farrington et al. (1980) and Neff (1979) have recently reviewed the literature on hydrocarbons in molluscs. A few systematic regional and local studies have been made of hydrocarbon concentrations in molluscs. Those studies dealing with U.S. coastal areas have focused primarily on the aromatic hydrocarbon benzo-a-pyrene, and occasionally a few other individual aromatic hydrocarbons. With the exception of samples from areas impacted by large oil spills, the concentrations of hydrocarbons are within the ranges of the concentrations in our data set. However, comparison of different stations is difficult because the methodology used to analyze for petroleum and fossil fuel hydrocarbons is not uniform and intercalibration of results is not routine, although there have been recent improvements (Wise et al., 1980; Rogerson et al., submitted).

A variety of parameters are used by various authors to estimate total petroleum contamination. There are no methods available to measure all petro-

leum compounds because of the vast number and wide molecular weight range of these compounds (Farrington et al., 1980). Our data demonstrates conclusively, as previously discussed and shown in Tables 10 and 13, that each of the parameters used, e.g. n-alkanes, phytane, UCM, total aromatics, can give a different indication of relative and absolute levels of petroleum contamination. Thus, we can only generally state that mussels and oysters collected close to urban and industrial centers in the world's coastal areas had elevated concentrations of fossil fuel compounds when compared to remote areas and the absolute differences in concentrations from one region or country to another is difficult to establish.

An example of the value of studies within a given region has been shown by the work of Burns and co-workers for Port Philip Bay and Westernport Bay, Australia where measurement of hydrocarbons in mussels was used to provide information on relative importance of various inputs of fossil fuel compounds to these marine ecosystems (Smith and Burns, 1978; Burns and Smith, 1978; Burns and Smith, 1981). An effort to characterize the pentacyclic triterpane composition of petroleum mixtures accumulated by mussels has begun in California in an attempt to determine contamination sources (Risebrough et al., in preparation).

Our data for hydrocarbons suggest that near urban areas there is a substantial input of petroleum hydrocarbons which are available for biological uptake. In addition, samples near urban areas also have relatively higher levels of pyrogenic hydrocarbons compared to more remote areas. The good agreement between 1976 and 1977 data for fl UCM and aromatic hydrocarbons suggests that the inputs to areas sampled are fairly constant. The Narragansett

Bay data that show month to month variations could be due to either or both environmental and biological controls on relatively constant inputs to a given area as well as fluctuating inputs of hydrocarbons. We cannot discern which of these mechanisms, if any, are dominant and can easily envision a simultaneous combination of processes that produces the net result that we measure. Also, we cannot tell if the origin of the petroleum hydrocarbons are repeated small spills or effluent discharges, or both. Either or both are reasonable sources based on our current limited knowledge of relative magnitudes of inputs (NAS, 1975; NOAA, 1979). A more detailed discussion of the Narragansett Bay data will be presented elsewhere after further investigations using sample collections for 1979 and 1980.

Thus far most analyses of petroleum and fossil fuel compounds in the environment have focused on n-alkanes, GC resolved branched alkanes, and aromatic hydrocarbons. The UCM of alkanes and cycloalkanes in the saturated hydrocarbon fraction and aromatic compounds in the aromatic fraction seem to survive the longest when petroleum is spilled into the environment. As we have stated elsewhere (Anderlini et al., 1980; Risebrough et al., 1980; Farrington et al., 1982), many cyclic alkanes such as diterpenoid and triterpenoid cycloalkanes and steranes are now more easily analyzed by computerized GC/MS and could prove to be the most stable long term markers of petroleum input and may even function as molecular signatures of certain specific inputs (Risebrough et al., 1980).

We have not presented or discussed in detail the large amount of biogenic hydrocarbon data which we have from the mussel and oyster analyses. This will be presented in a later paper. However, we do point out here that biogenic

hydrocarbons have been postulated as markers for marine food webs for many years (Blumer et al., 1964; review by Farrington and Meyers, 1975). We have the opportunity with the Mussel Watch data to test this hypothesis by determining if biogenic hydrocarbons in mussels reflect the expected biogenic hydrocarbons in phytoplankton and benthic algae growing in their habitat.

Other Pollutants

We have presented data for only a limited number of the many pollutant organic compounds known or suspected to be in the marine environment (NOAA, 1977). Our gas chromatograms obtained via EC detectors reveal many other halogenated compounds in addition to DDT family compounds and PCBs. Some of these peaks may represent biogenic brominated hydrocarbons or other naturally occurring compounds (Scheuer, 1978). However, many represent other pollutant chlorinated pesticides and industrial compounds. Flame ionization detector gas chromatograms and GC/MS computer systems analyses reveal many more compounds which are known or suspected pollutants. Several may be compounds as yet unrecognized as pollutants because they are by-products of manufacturing processes or they are unsuspected reaction products of other anthropogenic compounds released to the environment. One goal for the continuing "Mussel Watch" activities is to provide a means for rapidly searching existing GC and GC/MS data files to identify widely distributed ubiquitously disturbed anthropogenic compounds or pinpointing a geographic area with anamolously high concentrations of a given compound. We seek to maximize the detection of kepone type incidents (Hugget et al., 1980) in the early stages and think that the "Mussel Watch" concept offers one means of avoiding severe pollution problems of this type in the future.

We are conducting various statistical analyses of the present data set to discern further relationships between portions of the data which can provide insight to the biogeochemistry of pollutant compounds in estuarine and coastal areas. This will also assist in framing procedures for evaluating and interpreting the more complicated sets of data of the next several years.

Applications of "Mussel Watch" Data

We are strong proponents of the view that the "Mussel Watch" concept cannot and must not be viewed as a panacea for monitoring organic pollutants in the estuarine and coastal marine environment. It is only one approach among many which will provide a better understanding of the sources and rates of inputs, routes and rates of movement of pollutants to and through the marine environment. These measurements must be coupled with the full range of carefully designed field measurements of pollutants in other segments of the marine environment and experimentation with laboratory or larger scale systems.

For example, measurements in mussels provided a means of identifying the Achusnet River Estuary (New Bedford, and Fairhaven, Massachusetts) as a severe "hot spot" of PCB pollution but it was not possible to predict from these measurements the exact extent of PCB contamination of fish and lobsters in the area nor to assess the size of the sediment reservoir of PCBs in the harbor.

The "Mussel Watch" data can assist in providing an assessment of the extent of contamination of a given area from acute scale pollution events by providing both before and after measurements. For example, we had data for hydrocarbons in Mytilus edulis at the Cape Cod Canal, Massachusetts for 1976 and 1977 when a small oil spill occurred in 1978. This provided us with background measurements for comparison with measurements during and after the small

spill (Farrington et al., 1982). We have also shown that several oyster populations along the Gulf Coast, in particular the Texas coast which received oil input from the 1979 IXTOC-I oil well blowout (NOAA, 1981; Amos, 1980, among others), were already contaminated by petroleum hydrocarbons in 1976, 1977 and 1978 (Tables 10 and 18). This data provides background for assessing the extent and severity of petroleum contamination by an event such as the IXTOC-I oil spill.

The continuing measurements of chemical pollutants in mussels should provide an assessment of the ultimate fate of and at what rate DDT family compounds and PCB concentrations in the coastal environment decrease due to restriction on manufacture and use. This information is of value in first order predictions of environmental fates for other chemical pollutants released accidentally or deliberately to the coastal and estuarine areas of the world. This is important not only for developed nations such as the United States, but also for developing nations where lessons painfully learned by developed nations should assist in wise management of coastal and estuarine resources.

The increasing recognition of chemical pollution problems on land have forced a reconsideration of ocean disposal for hazardous waste as an alternative to land based disposal options (Goldberg, 1981; NACOA, 1981). The debate has been joined with respect to these options (Kamlet, 1981) and wise decisions on the "assimilative capacity" of the marine environment for chemical wastes" (Goldberg, 1981; Kamlet, 1981) will require knowledge of the biogeochemistry of pollutant compounds in coastal and estuarine systems. "Mussel Watch" has begun to build a data base and will continue to contribute to this knowledge.

CONCLUSIONS

1) Elevated concentrations of PCBs and fossil fuel hydrocarbons are associated with urban areas around the U.S. coast. Some concentrations are one to two orders of magnitude higher compared to remote areas. The northeast "megapolis" of the U.S. coast clearly shows elevated concentrations of PCBs and fossil fuel hydrocarbons from the Chesapeake Bay area to Boston, Massachusetts.

2) Samples with elevated concentrations of aromatic hydrocarbons show the influence of oil spill or chronic oil inputs in addition to the pyrogenic source composition for aromatic hydrocarbons found in samples from stations with low concentrations and remote from urban areas.

3) No single chemical indicator of fossil fuel compound contamination in samples can be used to predict the relative magnitude of other chemical indicators of fossil fuel contamination.

4) Monthly temporal changes of factors of two to ten are seen for the organic pollutants in mussels from the Narragansett Bay station, the only relatively contaminated station sampled at monthly intervals during the first phase of Mussel Watch.

5) Resampling and reanalysis of several stations - 1976, 1977, 1978 gave essentially the same results for DDT family compounds, PCBs, and fossil fuel hydrocarbons.

6) DDE concentrations for U.S. coastal areas in general were lower in 1976, 1977 and 1978 than in 1971 the last time measurements of DDE were made in shellfish sampled on this scale of geographical coverage.

7) The "Mussel Watch" concept works for detecting differences of an order of magnitude or greater between stations in different areas.

8) The "Mussel Watch" concept should work for less marked differences between stations of factors of 2 to 4 with refinements in analytical methods and better knowledge of the interactions between mussels and oysters and their habitat and the biochemistry of organic pollutants taken up by mussels and oysters.

9) "Mussel Watch" type measurement programs are only one type of the research and monitoring programs needed to provide knowledge of the fate and effects of chemical pollutants in the marine environment.

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FIGURE LEGENDS

Figure 1. U.S. EPA "Mussel Watch" sampling loations - 1976.

Figure 2. Glass capillary gas chromatograms of fraction 1 hydrocarbons (alkanes, cycloalkanes, alkenes, cycloalkenes) in Narragansett Bay monthly samples of Mytilus edulis for 1976. The numbers 12, 16, etc., indicate position of elution for n-C_{12}, n-C_{16} alkanes.

Figure 3. Glass capillary gas chromatograms of fraction 2 hydrocarbons (alkenes, cycloalkenes, aromatic hydrocarbons) for Narragansett Bay monthly samples of Mytilus edulis for 1976. The numbers 12, 16, etc., indicate position of elution for n-C_{12}, n-C_{16} alkanes.

Figure 4. Plot of 1976 vs. 1977 PCB concentration data for U.S. EPA "Mussel Watch" samples.

Figure 5. Woods Hole Oceanographic Institution Sea Grant and "Mussel Watch" stations for Massachusetts.

Figure 6. Representative glass capillary gas chromatograms of fraction 1 + fraction 2 hydrocarbons from a relatively heavily polluted station at Manhasset Neck, New York and Boston, Massachusetts and relatively pristine station at Blue Hills Falls, Maine. The numbers 12, 16, etc., indicate position of elution for n-C_{12}, n-C_{16} alkanes.

Figure 7. Bar graph presentation of aromatic hydrocarbon concentrations in samples of oysters from Sapelo Island, Georgia and Biloxi, Mississippi and mussels from Plymouth, Massachusetts and Manhasset Neck, New York.

CF
WB
GH
CR
T
Y
CB
GB
PSG
H
PA
BH
NSF
SSF
FAR
SC
SOB
SS
D
PTA
RC
SPH
SC
PF
OS
LJ
SDH
PAN
FUL
GAL
EMB
MAT
SAB
CAL
DEW
BAR
ANT
ESP
ILE
BAS
BRT
BOR
PAS
BIL
DAU
PEN
PAN
CAM
BLA
AP
KEY
AN
NAP
ST. A
SAV
SAP
CHR
FR
BU
H
LH
WW
CC
A
MN
HP
RK
NH
MIL
B
AC
MAY
CH
FI
P
CA
BI
PL
CCC
SK
BLK
GGI
SI
CN
BHF

Figure 1

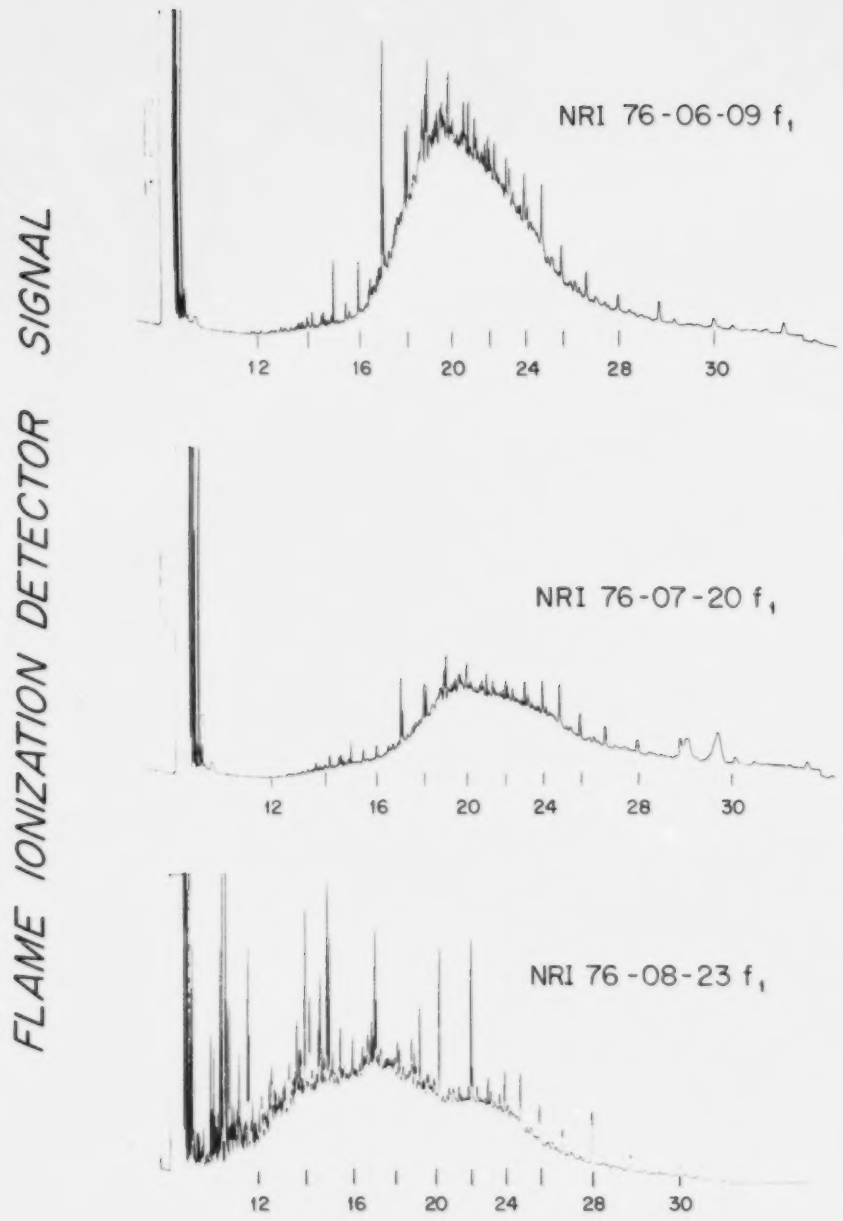
FLAME IONIZATION DETECTOR SIGNAL
NRI 76-06-09 f,
12
16
20
24
28
30
NRI 76-07-20 f,
12
16
20
24
28
30
NRI 76-08-23 f,
12
16
20
24
28
30

Figure 2a

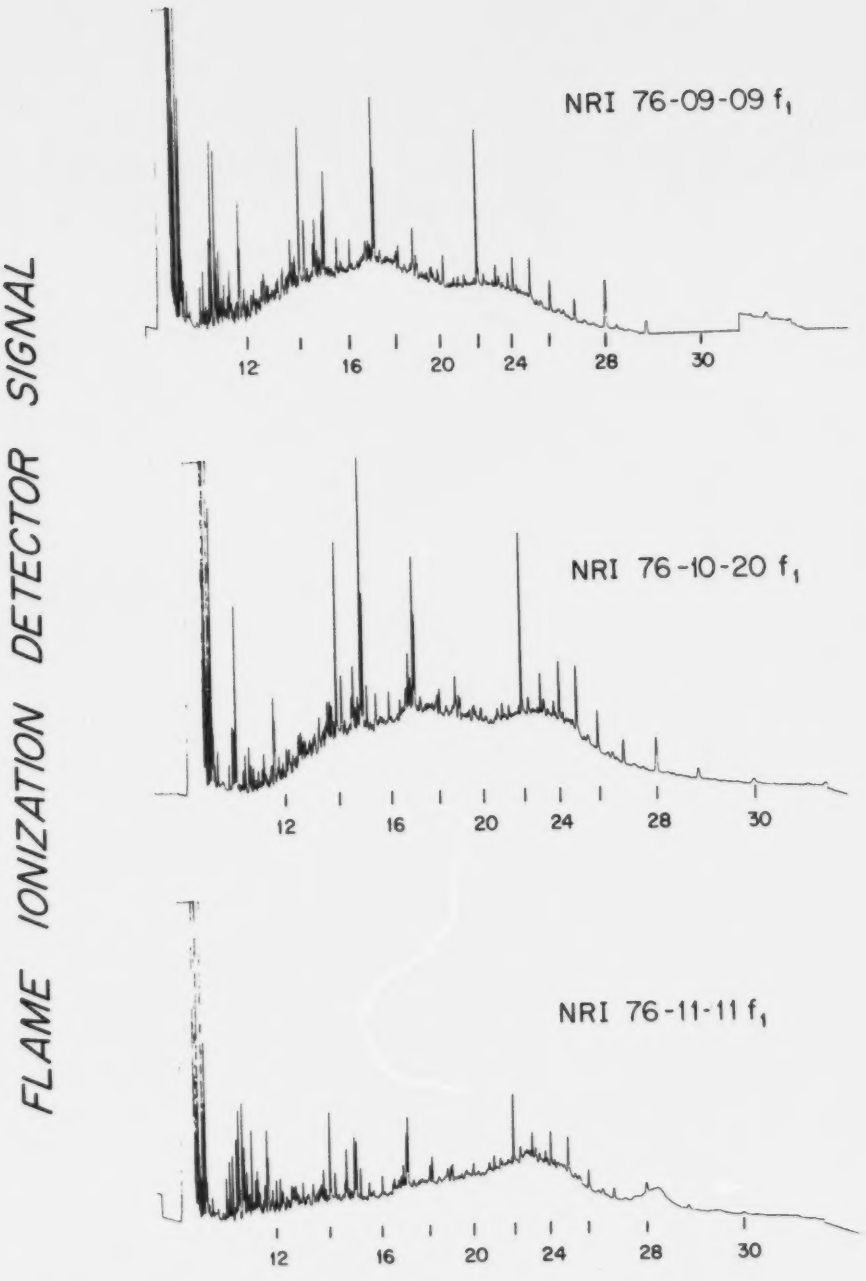
FLAME IONIZATION DETECTOR SIGNAL
NRI 76-09-09 f1
NRI 76-10-20 f1
NRI 76-11-11 f1
12
16
20
24
28
30

Figure 2b

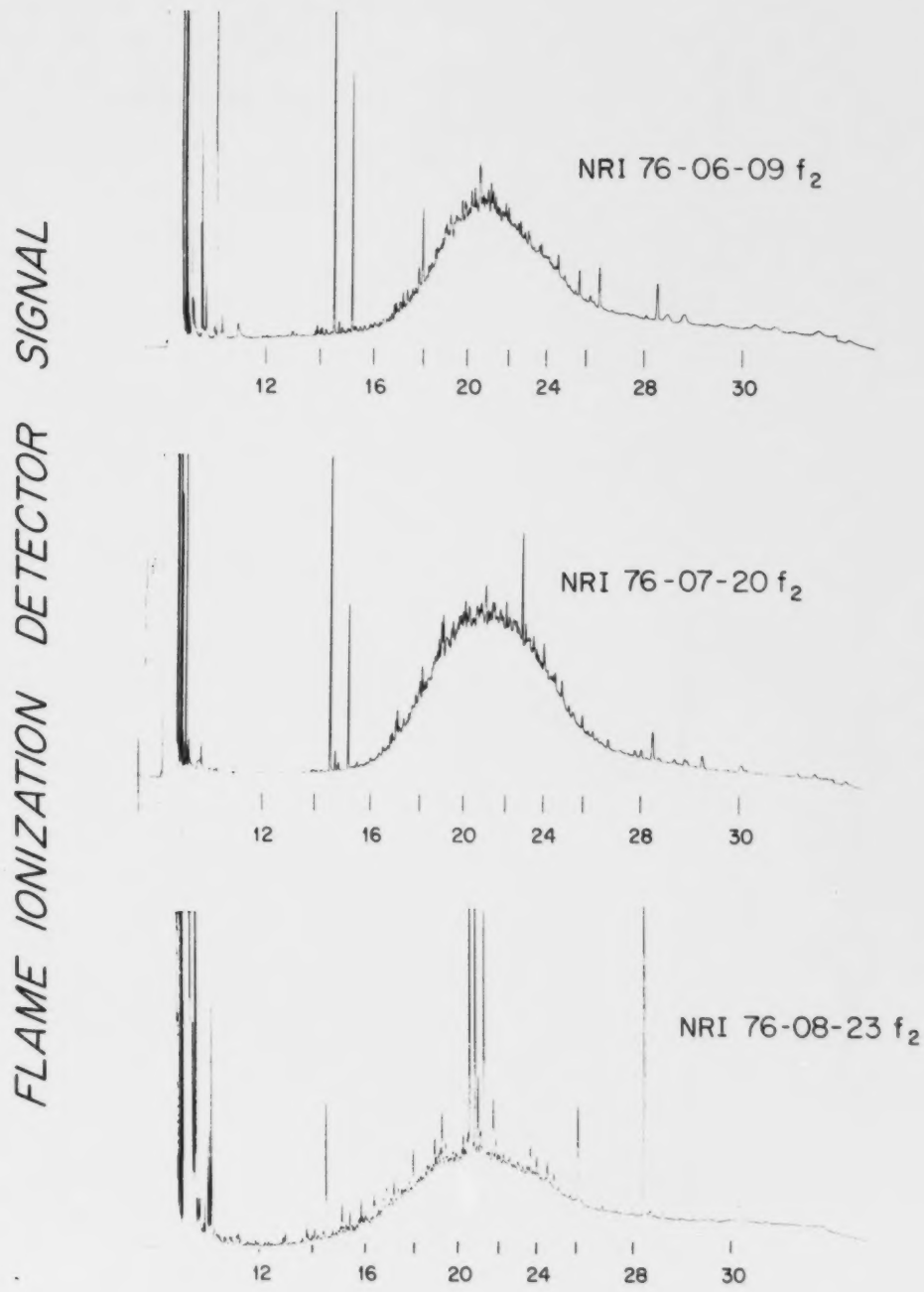
FLAME IONIZATION DETECTOR SIGNAL
NRI 76-06-09 f2
NRI 76-07-20 f2
NRI 76-08-23 f2
12
16
20
24
28
30

Figure 3a

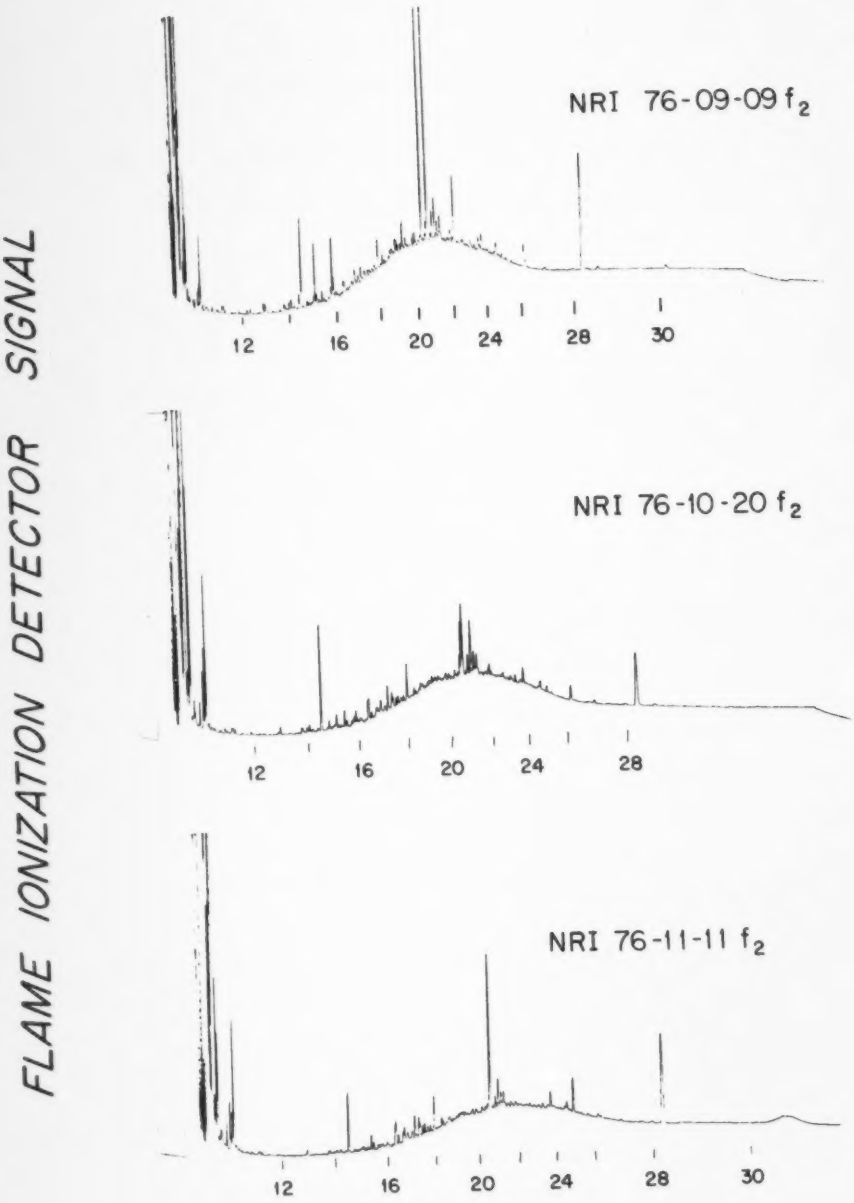
FLAME IONIZATION DETECTOR SIGNAL
NRI 76-09-09 f2
12
16
20
24
28
30
NRI 76-10-20 f2
12
16
20
24
28
NRI 76-11-11 f2
12
16
20
24
28
30

Figure 3b

U.S. MUSSEL WATCH PROGRAM

EAST COAST, GULF COAST AND WEST COAST

MUSSELS AND OYSTERS

PCB DATA 1976 - 1977

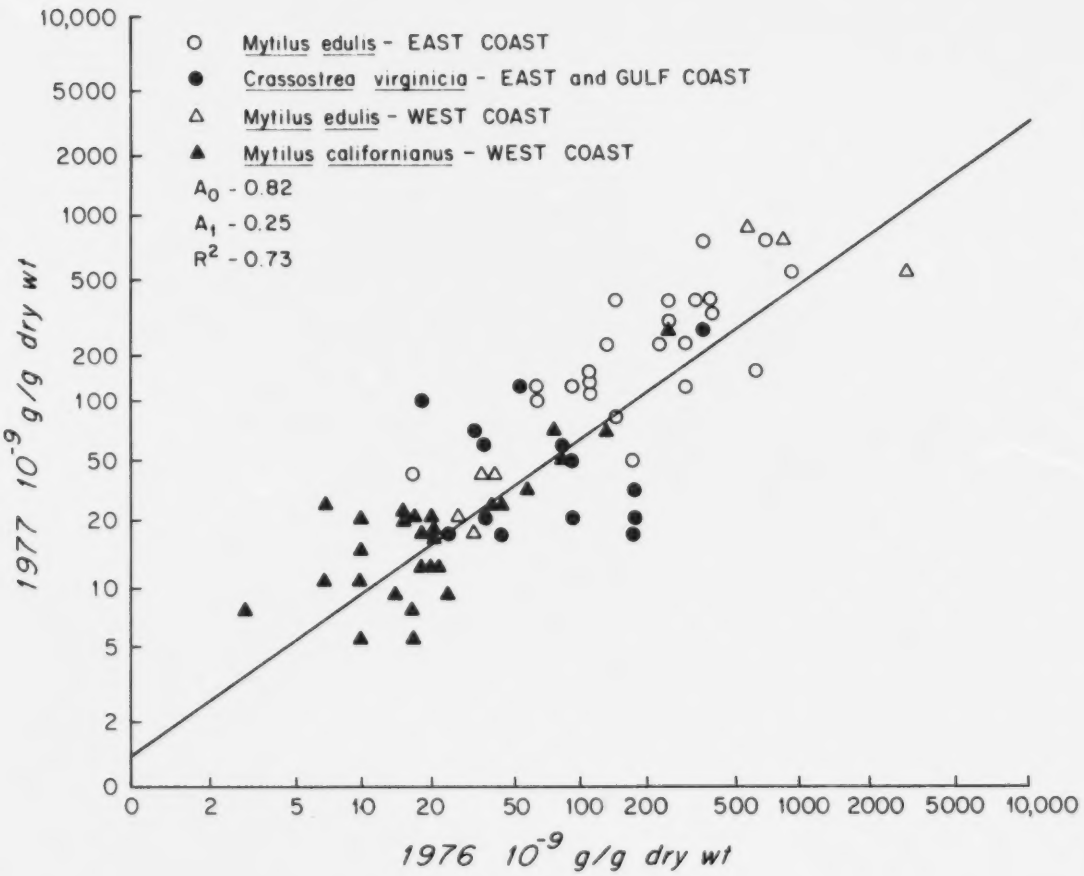

Figure 4

CAPE ANN
BOSTON
PLYMOUTH
CAPE COD CANAL
NEW BEDFORD
CUTTYHUNK
HARBOR
WEST POND
GAY HEAD
NANTUCKET
ESTHER IS
JETTY

Figure 5

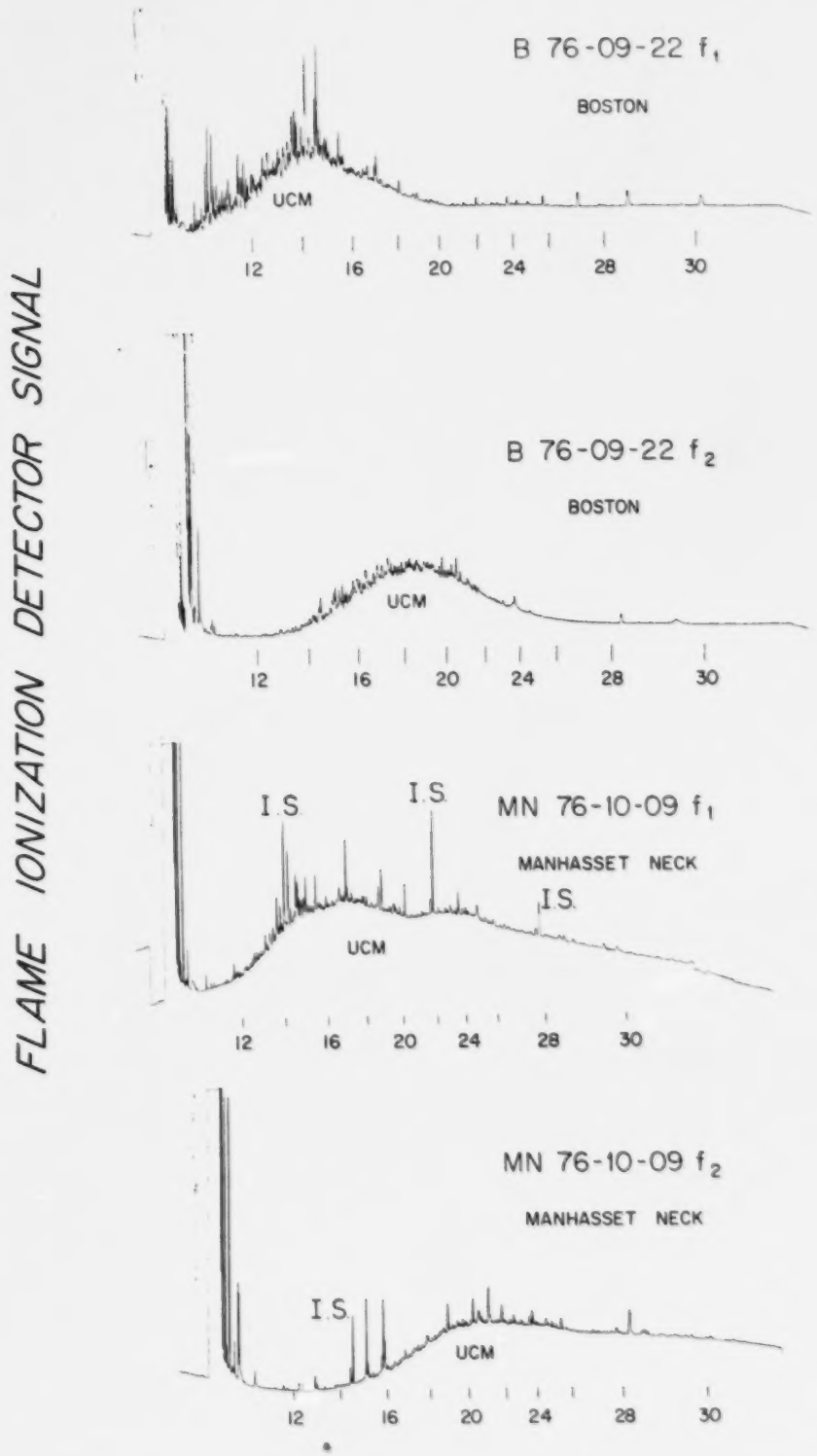
FLAME IONIZATION DETECTOR SIGNAL
B 76-09-22 f1
BOSTON
UCM
12
16
20
24
28
30
B 76-09-22 f2
BOSTON
UCM
12
16
20
24
28
30
I.S.
I.S.
MN 76-10-09 f1
MANHASSET NECK
I.S.
UCM
12
16
20
24
28
30
MN 76-10-09 f2
MANHASSET NECK
I.S.
UCM
12
16
20
24
28
30

Figure 6a

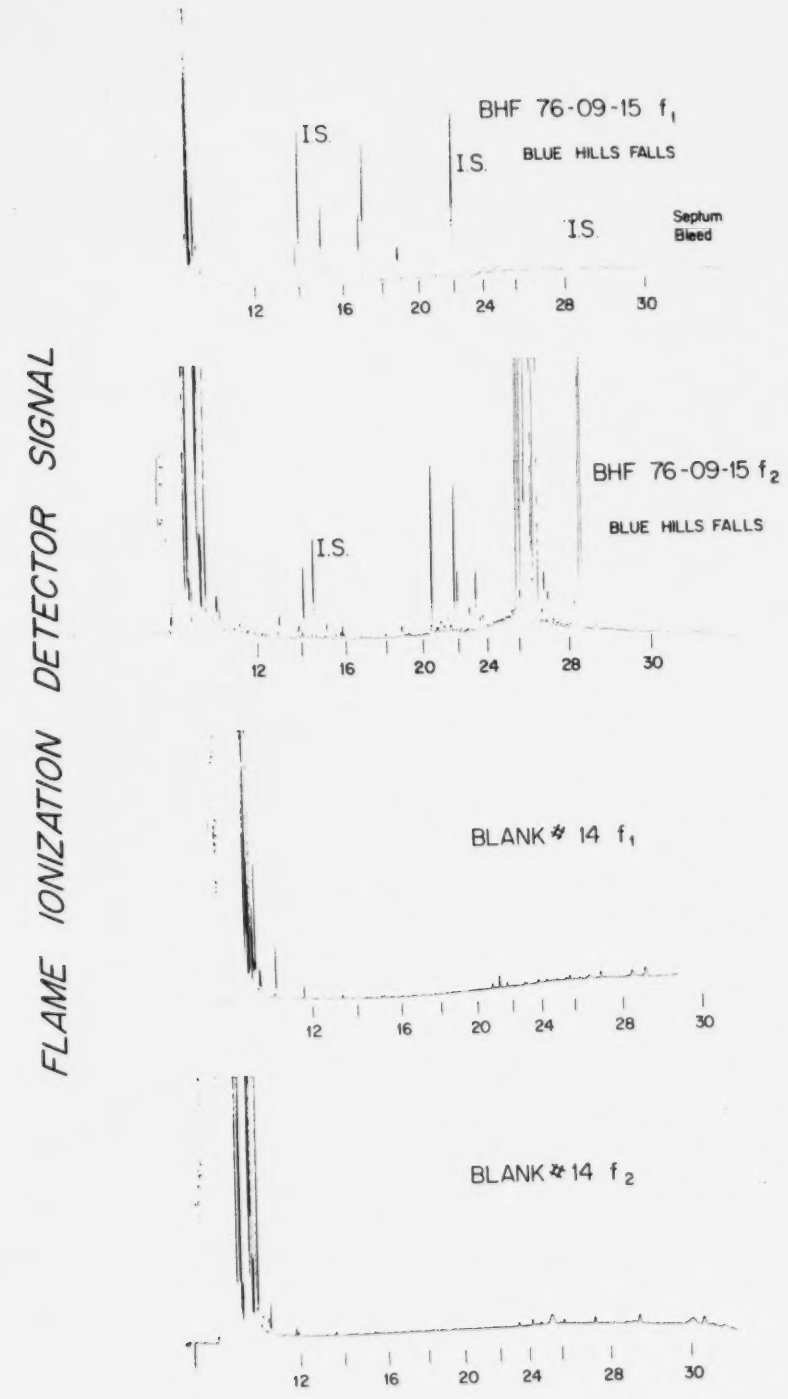
FLAME IONIZATION DETECTOR SIGNAL
BHF 76-09-15 f1
BLUE HILLS FALLS
I.S.
I.S.
I.S.
Septum Bleed
12
16
20
24
28
30
BHF 76-09-15 f2
BLUE HILLS FALLS
I.S.
BLANK # 14 f1
BLANK # 14 f2

Figure 6b

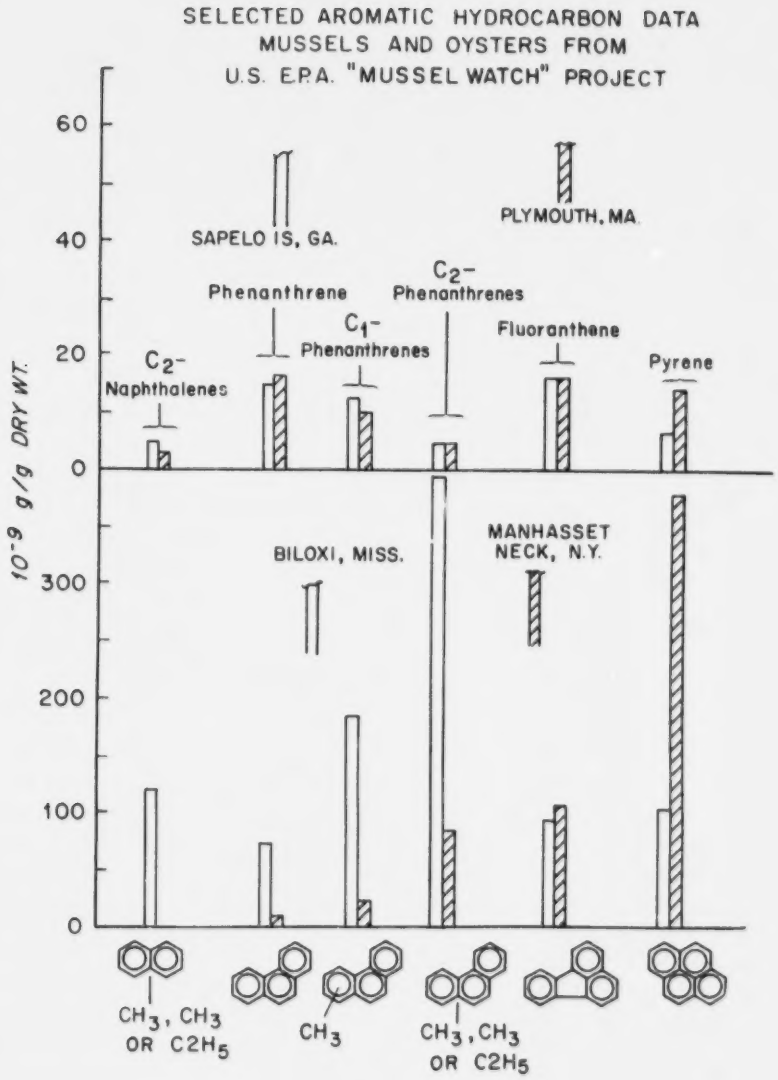
SELECTED AROMATIC HYDROCARBON DATA
MUSSELS AND OYSTERS FROM
U.S. E.P.A. "MUSSEL WATCH" PROJECT
10⁻⁹ g/g DRY WT.
60
40
20
0
300
200
100
0
SAPELO IS, GA.
PLYMOUTH, MA.
Phenanthrene
C2- Phenanthrenes
C1- Phenanthrenes
Fluoranthene
Pyrene
C2- Naphthalenes
BILOXI, MISS.
MANHASSET NECK, N.Y.
CH3, CH3 OR C2H5
CH3
CH3, CH3 OR C2H5

Figure 7

N O T E

FOR TABLES 3-19

DATA NOT CORRECTED FOR RECOVERY AND ARE PRECISE TO ONLY

TWO OR THREE SIGNIFICANT FIGURES DEPENDING ON

COMPOUNDS AND CONCENTRATIONS MEASURED.

TABLE 1

Station Locations.

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
Bodega Head	BH 760324	38°18.4'N 123°03.8'W	M. Californianus	Horizontal tidal ledge, upper range
Pt. La Jolla	LJ 760420	32°51'N 117°16.6'W	M. Californianus	Horizontal tidal ledge, upper middle range
San Diego Harbor	SDH 760425	32°40.9'N 117°14.2'W	M. edulis (1,2,3,5) M. Californianus (4)[e]	1 & 2 from concrete bridge standards 3, N. tip of Harbor Island 4, Point Loma, just inside harbor on granite breakwater 5, North Island on abandoned berths
Point Fermin	PF 760506	33°42.9'N 118°19'W	M. Californianus	Horizontal tidal ledge, upper reaches of mussel range
Ricon Cliffs	RC 670507	34°21.3'N 119°26.5'W	M. Californianus 15°C	Breakwater rocks, base of pier
Point Arguello	PTA 760508	34°34.7'N 120°38.8'W	M. Californianus	Tip of Arguello, N. side
San Pedro Harbor	SPH 760516	33°42.8'N 118°16.4'W	M. edulis	1, rock and concrete landfill 2, large granite rocks, base of pier, at entrance to Middle Harbor 3, concrete pier pilings 4, concrete pilings, Terminal Island side of Main Channel 5, Terminal Island side of Main Channel, below bridge and near berth
Oceanside jetty	OS 760523	33°11.6'N 117°23.4'W	M. Californianus	Granite beach jetty

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
Santa Catalina Is.	SCI 760604	33°27.1'N 118°29.2'W	M. Californianus	
San Simeon	SS 760614	35°37.2'N 121°09.2'W	M. Californianus	Horizontal tidal ledge
Diablo Canyon	D 760616	35°12.1'N 120°50.6'W	M. Californianus	Horizontal rock surface, upper range of mussels
Santa Cruz	SCZ 760622	37°57.1'N 122°04.9'W	M. Californianus	Horizontal tidal ledge, eroding sedimentary rock
Soberanes Pt.	SOB 760623	36°27'N 122°55.7'W	M. Californianus	Granite semi-horizontal ledge, upper-mid range
N. San Francisco (11 stations)	NSF 760701 (1)	38°3.8'N 122°13.5'W	M. edulis	Concrete pilings
	(2)	38°3.4'N 122°15.8'W	M. edulis	Concrete pilings
	(3)	38°3.3'N 122°18.3'W	M. edulis	Metal buoy
	(4)	38°2.4'N 122°20.9'W	M. edulis	Metal buoy
	(5)	38°0.7'N 122°24.1'W	M. edulis	Metal buoy
	(6)	37°57.8'N 122°25.9'W	M. edulis	Creosote wood pier pilings
	(7)	37°56.1'N 122°26.5'W	M. edulis	Creosote wood pier pilings
	(8)	37°51.8'N 122°23.7'W	M. edulis	Metal buoy
	(9)	37°49.9'N 122°28.7'W	M. edulis	Creosote pylon
	(10)	37°50.9'N 122°21.5'W	M. edulis	End of pier, 1 m above water level
	(11)	37°49.1'N 122°20.7'W	M. edulis	Concrete pilings, 1 m above water level

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
San Francisco (9 stations - continuation of NSF 760701)	SSF 760701 (12)	37°45.7'N 122°22.7'W	M. edulis	Creosote pier piling
	(13)	37°43.6'N 122°20.4'W	M. edulis	Wood substrate (apparently not creosote)
	(14)		M. edulis	Concrete piling
	(15)		M. edulis	Channel marker
	(16)		M. edulis	Channel marker
	(17)		M. edulis	Creosote wood pylon, main channel
	(18)		M. edulis	Channel marker
	(19)		M. edulis	Channel marker
	(20)		M. edulis	Concrete piling
Farallon Islands	FAR 760705	37°41.8'N 122°59.9'W	M. Californianus	Horizontal tidal ledge, natural rock, upper range
Point Arena	PA 760710	38°57.4'N 123°44.4'W	M. Californianus	Erosional sedimentary rock, upper range
Cape Mendocino	CM 760712	40°24.3'N 124°23.4'W	M. Californianus	Small rocky point, on soft conglomerate rock
Humboldt	H 760714	40°45.8'N 124°14.4'W	M. Californianus	On jetty rock
Point St. George	PSG 760716	41°46.7'N 124°15.2'W	M. Californianus	Natural rock (shale type), horizontal ledge, upper range
Gold Beach, Ore.	GB 760720	42°27.0'N 124°25.6'W	M. Californianus	Semi-horizontal natural rock
Coos Bay, Ore.	CB 760723	143°19.9'N 24°22.9'W	M. Californianus	Horizontal tidal shelf, natural rock
Yaquina Head	Y 760726	44°40.7'N 124°04.7'W	M. Californianus	On natural rock, upper ranges from small rocky point
Tillamook Bay (5 stations)	T 760729 (1)	45°31.5'N 123°53.8'W	M. edulis	Man-made rock fill-point, near oyster processing plant
	(2)	45°32.1'N 123°53.9'W	M. edulis	Flood channel, concrete substrate

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
Tillamook Bay (5 stations)	(3)	45°32.9'N 123°54.4'W	M. edulis	On concrete remnants
	(4)	45°33.5'N 123°55.1'W	M. edulis	Natural rock substrate
	(5)	45°33.9'N 123°56.2'W	M. edulis	Entrance channel to bay
Columbia River	CR 760802	46°14.0'N 124°03'W	M. Californianus	Jetty rock
Willapa Bay (2 stations)	WB 760804 (1)	46°40.4'N 123°55.6'W	M. edulis	Natural rock substrate, mussels are underneath rock at mud line
	(2)	46°43.7'N 124°04.0'W	M. edulis	Area largely mud-flats for both stations
Grays Harbor	GH 760806	46°55.7'N 124°10.5'W	M. Californianus	Area largely mud-flats, mussels from jetty
Cape Flattery	CF 760810	48°23.1'N 123°43.6'W	M. Californianus	Horizontal tidal ledge, natural rock substrate
Puget Sound	PS 760812	47°54.2'N 122°22.9'W	M. edulis	Natural rock, south end of Whidbey Island
Boundary Bay	BB 760818	48°56.5'N 122°49.2'W	M. edulis	Natural rock
Blue Hills Falls, Maine	BHF 760915	44°22.5'N 68°33.5'W	M. edulis	Horizontal tide-pool, upper range
Sears Island, Maine	SI 760916	44°25.8'N 68°53.1'W	M. edulis	Horizontal tide-flat, small rocky area
Cape Newagen, Maine	CN 760917	43°47.2'N 69°39.2'W	M. edulis	Natural rock
Bailey Island, Maine	BI 760918	43°44.9'N 69°59.4'W	M. edulis	Rock ledges
Portland, Maine	P 760920	43°41.2'N 70°13.9'W	M. edulis	Stone-boulder beach substrate
Cape Ann, Mass.	CA 760921	42°39.6'W 70°37.3'W	M. edulis	Tide-pool rocks, adjacent to sandy beach

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
Boston	B 760922	42°21.3'N 70°58.1'W	M. edulis	Small stone beach, northwest corner Deer Island
Plymouth, Mass.	PLY 760923	41°56.5'N 70°36.7'W	M. edulis	Rocky-stone beach
Cape Cod Canal, Mass.	CCC 760925	41°44.45'N 70°36.8'W	M. edulis	Granite rock along canal
Sakonnet Point, Rhode Island	SK 760927	41°27.2'N 71°11.7'W	M. edulis	Natural rock shelves, between sandy beaches
Block Island, Rhode Island	BLK 761001	41°11.6'N 71°35.7'W	M. edulis	Small stone beach
Millstone, Conn.	MIL 761004	41°18.3'N 72°09.8'W	M. edulis	Natural rock tidal ledges, S. of nuclear power-plant outfall (temp. 28°C near outfall)
New Haven, Conn.	NH 761006	41°11.5'N 73°03.2'W	M. edulis	Rock substrate, Charles Island
Manhasset Neck, New York	MN 761009	40°51.9'N 73°43.9'W	M. edulis	Shell-bed substrate on coarse sand to small rock beach
Herod Point, Long Island, New York	HP 761010	40°58.1'N 72°50.7'W	M. edulis	Stone-sand substrate beach
Fire Island, Long Island, New York	FI 761011	40°46'N 72°45.1'W	M. edulis	Jetty rock
Great Gull Island	GGI 761011	41°12.2'N 72°07.1'W	M. edulis	
Rockaway Point, New York	RK 761012	40°32.5'N 73°56.5'W	M. edulis	Jetty rock substrate
Atlantic City, New Jersey	AC 761014	39°22.3'N 74°24.3'W	M. edulis	Jetty rock substrate
Cape May, New Jersey	MAY 761015	38°55.6'N 74°57.8'W	M. edulis	Jetty rock substrate

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
Cape Henlopen, Delaware	CH 761104a	38°46.8'N 75°07.7'W	M. edulis	Jetty rock substrate
	CH 761104b	38°46.8'N 75°07.7'W	C. virginica	
Assateaque, Maryland	A 761106	38°19.4'N 75°05.6'W	M. edulis	Jetty rock
Cape Charles, Virginia	CC 761109	37°17.3'N 76°01'W	C. virginica	Tidal marshland, substrate is old oyster shells and mud
Lynnhaven Bay, Virginia	LH 761115	36°54.2'N 76°05.3'W	C. virginica	Marsh-grass habitat
Wachapreague Inlet, Virginia	W 76116a	37°35.2'N 75°36.8'W	M. edulis	Wood pier pilings
	W 76116b	37°35.2'N 75°37.8'W	C. virginica	
Hatteras Island, North Carolina	H 761119	35°12.3'N 75°43'W	C. virginica	Tidal marshland, mud substrate
Beauford, North Carolina	BU 761121	34°43'N 76°40.7'W	C. virginica	Tidal basin
Cape Fear, North Carolina	FR 761122	33°57'N 77°55.5'W	C. virginica	Tidal marshland
Sapelo Island, Georgia	SAP 761129	31°23.6'N 81°16.6'W	C. virginica	Tidal creek
Savannah River, Georgia	SAV 761130	32°01'N 80°52.4'W	C. virginica	Banks of South Channel
Charleston, South Carolina	CHR 761201	32°44.2'N 79°52.5'W	C. virginica	Fort Johnson
St. Augustine, Florida	STA 761204	29°41.5'N 81°13.8'W	C. virginica	Tidal marsh
Smithsonian Harbor Branch Laboratory	SMTH 761218		O. equestris	Raised in laboratory tanks
Naples, Florida	NAP 761223	26°01.3'N 81°44.1'W	O. equestris	Mangrove roots

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
Anclote, Florida	AN 761230	28°11.4'N 82°47.5'W	O. equestris	Oyster beds north of electric power-plant
Cedar Key, Florida	KEY 761231	29°03.5'N 82°59'W	C. virginica	Commercial oyster-bed[e]
Apalachicola, Florida	AP 770104	29°42.9'N 84°53.4'W	C. virginica	Commercial oyster-bed[e]
Panama City, Florida	PAN 770105	30°15'N 85°45'W	C. virginica	West Bay of St. Andres Bay[e]
Pensacola, Florida	PEN 770108	30°20.3'N 87°09.5'W	C. virginica	Sabine Point, Santa Rosa Island
Dauphin Island, Alabama	DAU 770113	30°18'N 88°08.5'W	C. virginica	Commercial subtidal reef[e]
Pass Christian, Mississippi	PAS 770115	30°18'N 89°15.8'W	C. virginica	Subtidal reef[e]
Biloxi, Mississippi	BIL 770117	30°23.4'N 88°51.5'W	C. virginica	Shallow waters, next to dock
Lake Borgne, Louisiana	BOR 770120	29°57.5'N 89°39.1'W	C. virginica	Southwest area of lake[e]
Lake Campo, Louisiana	CAM 770123	29°39.8'N 89°38.7'W	C. virginica	e
Black Bay, Louisiana	BLA 770124	29°37'N 89°37'W	C. virginica	West area of bay[e]
Bastian Bay	BAS 770125	29°19.2'N 89°38.9'W	C. virginica	Northern area of bay[e]
Bayou de West, Louisiana	DEW 770126	29°12'N 91°03.4'W	C. virginica	Between Bay Junop and Gulf of Mexico[e]
Lake Barre, Louisiana	BAR 770126	29°15.7'N 90°34.1'W	C. virginica	West side of lake[e]
Bay de Illettes, Louisiana	ILE 770127	29°17.1'N 90°00.1'W	C. virginica	In Bay de Illettes, just west of Point Peiro; oil-drilling in area[e]
Barataria Bay, Louisiana	BRT 770128	29°21'N 89°55.9'W	C. virginica	Central area of bay, near Middle Bank Light[e]

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
Calcasieu Lake, Louisiana	CAL 770201	29°50.4'N 93°20.2'W	C. virginica	South end of lake, just east of ship channel[e]
Galveston Bay	GAL 770219	29°29.2'N 94°46.3'W	C. virginica	
East Matagordo Bay	EMB 770315	28°40.0'N 95°55.1'W	C. virginica	
Fulton	FUL 77036	28°08.5'N 97°02.9'W	C. virginica	
San Antonio Bay	ANT 770316	28°18.6'N 96°40.2'W	C. virginica	
Panther Point	PAN 770316	28°12.5'N 96°42.0'W	C. virginica	
Matagordo Bay	MAT 770316	28°38.0'N 96°02.0'W	C. virginica	
Espiritu Santo Bay	ESB 770310	28°18.6'N 96°36.0'W	C. virginica	
Lake Sabine Bay	SAB 770318	29°43.2'N 93°51.5'W	C. virginica	

1977 Collections[d]
Special Notations
Location

NSF only substations 1, 2, 7, 9, 10, 11 collected
SSF only substations 12, 13, 14, 15, 16, 17 collected
WB only substation 2 collected

Stations Discontinued or not Collected - 1977

SMTH, BOR, CAM, BAS, DEW, BAR, ILE, BRT, FUL, PPT, ESB, GGI

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
New Stations				
Fort Pierce	FORP	27°29.1'N 80°18'W	C. virginica	
Pumpkin Bay	PUK	29°40.2'N 89°33.1'W	"	
Quarantine Bay	QRT	29°24.4'N 89°32.1'W	"	
Junop Bay	JUN	29°12.2'N 91°03.1'W	"	
Golden Meadows	GOD	29°15.1'N 90°19.1'W	"	
Drum Bay	DRM	29°52.8'N 89°20.0'W	"	
Brownsville	BRV	26°02.1'N 97°11.0'W	"	
Mesquite Bay	MEQ	28°08.2'N 96°50.1'W	"	
Lavaca Bay	LAV	28°36.1'N 96°33.4'W	"	
Lavaca Bay North	LAVN	28°40.1'N 96°35.1'W	"	
Aransas Bay	ARN	28°02.3'N 97°01.0'W	"	

1978 Collections[d] (includes 1976 and 1977 stations except as noted)

Stations Not Collected

WB, TIL substations 3 and 4, HI, GGI, PUK, JUN, GOD, DRM, EMB, BRV, ANT, MEQ, LAV, LAVN, BIL, PEN

(Cont.)

Location	Station Initials[a] & Date[b]	Fix	Species	Description of Site
New Stations				
New Bedford	NB	41°37.6'N 70°55.0'W	M. edulis	Rocky intertidal near hurricane dike
Tampa Bay	TAB	27°45'N 82°31'W	C. virginica	e.
California Bay	CAI	29°30.2'N 89°35.6'W	"	e.
Chaland Bay	CHL	29°14.5'N 90°38.3'W	"	e.
Terrabone Bay	TER	29°06'N 90°35'W	"	e.
Monthly Stations				
Narragansett Bay	NRI 7606-7608 7609-7611 7706-7812	41°31.3'N 71°19.6'W 41°28.5'N 71°23.9'W 41°38.5'N 71°22'W	M. edulis	Coddington Point; Intertidal rocks West Shore, Beavertail Neck's intertidal rocks West of Prudence Island; Subtidal, dredged
Bodega Head	BH	38°18.4'N 123°03.8'W	M. californianus	Rocky headland

[a]Initials of the station's name.
[b]Date of collection, starting with year, then month, then day. A small letter or seventh digit in some samples refers to the station sequence.
[c]Explanations are given in the right-hand column.
[d]Dates of collections given in data tables. Change in stations largely dictated by non-availability of species at earlier station locations. New stations added to replace old stations or to increase sampling density in given region.
[e]Samples and locations provided by local commercial fishermen and/or local natural resource officials.

TABLE 2

Synopsis of Total Data Set Entries for Most U.S. Mussel Watch Stations: PCB, DDE, and Hydrocarbon Analyses.

(1) Sample location code, date, bivalve type

(2) Numbers of individuals analyzed

(3) Range and average shell lengths

(4) Percentage tissue

(5) Percentage moisture

(6) Hydrocarbon concentrations

a. Total alkane + cycloalkane fraction
b. Total aromatic + alkene-cycloalkene fraction
c. 10-50 Individual n-alkane, branched alkane, alkene-cycloalkenes (major gas chromatogram peaks)
d. 5-10 Major aromatic hydrocarbons in molecular weight range of naphthalene to fluoranthene and pyrene

(7) PCB concentrations

(8) DDT family compounds - mainly DDE

TABLE 3

PCB and DDE Concentrations in Bodega Head Mussels
(Mytilus californianus)
(10^{-9} g/g dry weight).

Date	DDE				PCB (as 1254 mixture)			
	B.M.L.[i]	W.H.O.I.[ii] Duplicates			B.L.M.	W.H.O.I. Duplicates		
		a[ii]	b	Avg.		a	b	Avg.
760324	17	34	35	35	10	36	67[iv]	52
760506	18	--[iii]	--	--	22	--	--	--
760603	10	20	21	21	19	17	35[iv]	26
760702	15	--	--	--	20	--	--	--
760729	23	15	13	14	16	27	97[iv]	62
760910	8	--	--	--	10	--	--	--
761006	7	13	15	14	18	20	16	18
761202	--	33	36	35	--	20	22	21
770104	4.6	19	17	18	27	17	16	17
770202	24	19	21	20	34	25	34	30
770303	15	14	15	15	21	14	15	15
770408	14	--	--	--	24	--	--	--
770506	9	--	--	--	13	--	--	--
770607	7	--	--	--	18	--	--	--
770705	12	--	--	--	13	--	--	--
770803	11	--	--	--	21	--	--	--
770831	13	--	--	--	11	--	--	--
770930	5	--	--	--	4.8	--	--	--
771108	4	--	--	--	18	--	--	--
771208	6.3	--	--	--	20	--	--	--

[i] B.M.L. = Bodega Marine Laboratory - Risebrough et al.;
W.H.O.I. = Woods Hole Oceanographic Institution - Farrington et al.

[ii] Duplicate analyses of subsamples of homogenate of 20-30 mussels.

[iii] Not analyzed.

[iv] Poor reproducibility.

TABLE 4

PCB Concentrations (as 1254 PCB mixture) Narragansett Bay, Rhode Island Mussels (Mytilus edulis)[i] (10^{-9} g/g dry weight).

Date	Duplicates[ii]		Avg.
760609	568	685	626
760720	535	457	496
760823	332	230	281
760909	465	485	475
761020	297	302	300
761111	347	395	371
770623	341	404	373
770718	346	339	342
770810	345	313	329
770914	237	234	236
771017	365	310	338
771118	397	387	392
771212	358	314	336
780222	703	567	635
780428	459	408	434
780523	370	333	352
780630	446	461	454
780721	519	540	530
780829	607	595	601
781020	398	409	404
781121	321	284	302
781228	451	374	412

Narragansett "Mega" Mussel Intercalibration, 1979

Bodega Marine Laboratory (10^{-9} g/g dry weight)	Woods Hole Oceanographic Institution (10^{-9} g/g dry weight)
580 ± 23 (packed GC column) (n=3)	412 ± 23[iii] (n=4)
510 ± 140 (capillary GC column) (n=3)	

[i] Woods Hole Oceanographic Institution - Farrington et al.

[ii] Duplicate analyses of subsamples of homogenate of 20 to 50 mussels.

[iii] Not corrected for 80% recovery.

TABLE 5

Texas Oyster Intercalibration Sample: Hydrocarbon Analyses.

(10^{-6} g/g dry weight).

Retention Index[ii]	U.T.[i] Subsamples 1	U.T.[i] Subsamples 2[iii]	W.H.O.I.[i] Subsamples 1[iii]	W.H.O.I.[i] Subsamples 2[iii]
15.80	.09	.33	--	--
16.00	.30	.28	.24	.33
16.60	.65	.84	.89	1.2
17.00	.26	.28	.44	.52
17.70	.81	.98	.960	1.2
18.00	.36	.42	--	--
18.50	.43	.36	.37	.45
19.00	.31	.40	.37	.45
19.50	.20	.10	--	--
19.60	.13	.08	.19	.28
20.00	.15	.24	.24	.35
20.70	.07	.17	.11	.20
21.00	.23	.26	.37	.53
22.00	.23	.46	.40	--
22.60	.25	.24	--	--
23.00	.28	.44	.52	.67
23.60	.20	.14	--	--
24.00	.26	.33	.52	.80
24.60	.07	.07	--	--
25.00	.26	.30	.41	.52
26.00	.21	.23	.34	.50
27.00	.19	.20	.28	.38
28.00	.22	.31	.24	--
29.00	.25	.53	.25	.44
30.00	.27	.69	.17	.33
31.00	.33	.90	.13	.28
32.00	.35	.89	.09	.19
33.00	.33	.82	.07	.14
Total	7.69	11.31	7.61	9.83
Total UCM	--	--	120	131

[i] U.T. = University of Texas Marine Science Institute, Port Aransas - Parker et al.; W.H.O.I. = Woods Hole Oceanographic Institution - Farrington et al.

[ii] Retention time compared to nearest eluting standard hydrocarbon, e.g. 1600 is n-hexadecane, 1700 is n-heptadecane.

Note: Differences in values and in retention index may be due to differences in chromatographic columns used in the analysis.

[iii] Subsamples of oyster homogenate prepared from oysters contaminated during an accidental oil spill on the coast of Texas.

TABLE 6

Intercomparison of W.H.O.I. and University of Texas Analyses of Alkanes in NBS Intercalibration Mussels (10^{-9} g/g dry weight).

Compound	Alaska Mussels				Santa Barbara Mussels			
	W.H.O.I.[i]			U.T.[i]	W.H.O.I.[i]			U.T.[i]
	# of Analyses	A13 + A44[ii] Range	Average	A11[ii]	# of Analyses	S2 + S47[ii] Range	Average	S48[ii]
n-C_{14}	3[iii]	109 - 154	136	180	3	83 - 119	104	137
n-C_{16}	3	155 - 199	178	446	3	100 - 201	146	491
n-C_{18}	3	22 - 28	25	475	3	65 - 166	112	334
n-C_{20}	2	20 - 21	21	257	3	66 - 159	110	266
n-C_{22}	1	88	88	558	2	138 - 221	180	388
n-C_{24}	3	13 - 68	37	347	4	86 - 221	140	684
n-C_{25}	4	32 - 151	59	217	4	132 - 257	192	1717
n-C_{27}	4	24 - 88	45	299		N.C.[iv]	N.C.	1606
n-C_{28}	2	39 - 78	59	327		N.C.	N.C.	N.C.
Pristane	3	1490 - 2340	1957	1983	3	247 - 498	369	478
Phytane	3	39 - 57	49	28	3	120 - 237	180	43

[i] U.T. = University of Texas, Port Aransas - Parker et al.;
W.H.O.I. = Woods Hole Oceanographic Institution - Farrington et al.

[ii] Subsamples received from U.S. NBS (see Wise et al., 1980).

[iii] Several times compounds in sample could not be calculated due to spike of compounds to calculate recoveries. This is reflected in number of analyses used to measure the concentration of the compound.

[iv] N.C. = Not calculated as peak was too small or poorly resolved.

TABLE 7

Representative Duplicate Analyses for n-Alkanes, Branched Alkanes, and Unresolved Complex Mixture (UCM) Alkanes Plus Cycloalkanes

(10^{-9} g/g dry weight individual compounds)

(10^{-6} g/g dry weight UCM).

Compound or GC Retention Index	BHF 760915(e)[i]		B 760922(e)		SAV 761130(0)		BIL 770117	
14.2[iii]	--[ii]	--	19×10^{2}	23×10^{2}	12×10^{1}	12×10^{1}	16×10^{1}	14×10^{1}
14.7	13	17	20×10^{2}	22×10^{2}	20×10^{1}	20×10^{1}	83×10^{1}	83×10^{1}
14.9	18	22	38×10^{1}	35×10^{1}	75	73	--	--
15.0	12×10^{1}	18×10^{1}	18×10^{1}	23×10^{1}	26×10^{1}	25×10^{1}	21×10^{1}	22×10^{1}
15.1	18	46	13×10^{1}	87	57	60	--	--
16.0	24	49	11×10^{1}	13×10^{1}	93	91	11×10^{1}	12×10^{1}
16.9	77	17×10^{1}	--	--	--	--	--	--
17.0	13×10^{1}	33×10^{1}	32×10^{1}	36×10^{1}	26×10^{1}	25×10^{1}	21×10^{1}	19×10^{1}
Pristane	--	--	49×10^{1}	53×10^{1}	97	86	52×10^{1}	50×10^{1}
Phytane	--	--	33×10^{1}	34×10^{1}	60	59	48×10^{1}	48×10^{1}
21	--	--	--	--	--	--	--	--
23	27	14	12×10^{1}	66	--	--	--	--
25	56	21	27×10^{1}	31×10^{1}	--	--	--	--
UCM (10^{-6} g/g dry weight)	< 5	< 5	29×10^{1}	31×10^{1}	33	34	68	91

[i] See Table 1 for code.

[ii] None detected at 1.0×10^{-9} g/g detection limit.

[iii] Retention index of 15.0 is n-C_{15} (pentadecane); 14.2 refers to position of GC elution 2/10 time between elution of n-C_{14} (tetradecane) and n-C_{15}.

TABLE 8

Representative Duplicate Analyses for Aromatic Hydrocarbons in Mussels and Oysters: U.S. EPA Mussel Watch Stations.

(10^{-9} g/g dry weight).

Compound	SK 760927(e)[i]		BLK 761001(e)		W 761116(e)		BIL 770117(o)	
Naphthalene	--	--[ii]	--	--	8.2	4.4	5.6	4.1
Methylnaphthalene	--	--	.8	1.6	5.7	--	14	29
C_2 - Naphthalene	--	--	23	17	--	--	66	121
Phenanthrene	9.9	14	20	18	28	24	43	76
Methyl Phenanthrene	6.4	5.7	33	30	19	21	120	190
C_2 - Phenanthrene	--	--	41	44	8.4	3.8	258	396
Fluoranthene	12	14	37	40	23	17	42	94
Pyrene	3.9	5.2	23	22	12	10	63	104

[i]See Table 1 for sample code.

[ii]None detected at 0.1 x 10^{-9} g/g dry weight detection limit.

TABLE 9

Selected Hydrocarbon Concentrations in Bodega Head Mussels (Mytilus californianus) (10^{-6} g/g dry weight).

Sample Code	n-C_{17}[i]	2016[i]	2039[i]	2058[i]	2076[i]	2094[i]	Total[ii]
BH 760702	.62	--	--	--	.17	--	4.4
BH 760729	.055	.015	--	.16	--	--	.96
BH 760603	.022	--	.032	--	--	--	.98
BH 760910	--	--	--	.75	--	--	2.3
BH 761006	.13	--	.56	--	--	.45	1.8
BH 761104	.16	--	1.3	--	--	2.3	6.4
BH 761202	--	--	.029	--	--	.047	0.09
BH 770104	.57	.082	.054	.15	.064	.035	13
BH 770202	2.9	--	--	--	--	.70	27
BH 770303	.52	--	--	--	.28	.97	6.9
BH 770408	.098	--	--	--	.58	.14	2.3
BH 770506	.047	.25	--	--	--	--	2.9
BH 770607	.21	--	.019	--	.095	.29	9.1
BH 770705	.16	--	--	--	.11	--	6.2
BH 770803	.14	--	--	--	.057	--	3.2
BH 770831	.019	--	--	--	.023	--	1.1
BH 770930	.064	--	--	--	.075	--	4.1
BH 771108	.24	--	--	--	.72	.060	12
BH 771202	.20	--	--	--	.23	.036	6.0

[i] n-C_{17} - n-heptadecane; 2016, 2039, etc. refer to compounds with these retention indices whose exact structure is unknown but are of the alkene or cycloalkene type.

[ii] Total resolved peaks: alkanes.

TABLE 10

Selected Hydrocarbon Concentrations in Narragansett Bay Mussels (Mytilus edulis) (10^{-6} g/g dry weight) (duplicate measurements).

Sample Date[i]	n-C_{15}	n-C_{16}	n-C_{17}	Pristane	Phytane	UCM[iv]
760609	.045	.039	.18	.060	.060	165
	--[ii]	--[ii]	.097	.065	.059	105
760720	.017	.010	.050	.020	.024	86
	.107	.062	.091	.020	.017	79
760823	.29	.080	.29	.12	.060	80
	.33	.072	.30	.15	.090	82
	Station Location Change (See Text)					
760909	.21	.057	.36	.20	.055	67
	.22	.065	.34	.20	.057	62
761020	.34	.075	.41	.25	.070	69
	.32	.080	.42	.24	.067	80
761111	.26	.091	.25	.33	.13	101
	.25	.090	.27	.34	.14	88
	Station Location Change - (See Text)					
770623	.058	.038	.080	.025	N.D.[iii]	15
	.072	.047	.11	.027	N.D.	31
770718	.067	.034	.095	.034	N.D.	15
	.063	.031	.089	.010	N.D.	13
770810	.18	.074	.18	.036	N.D.	14
	.15	.062	.16	.030	N.D.	15
770914	.071	.036	.12	.009	N.D.	8
	.066	.041	.12	.013	N.D.	7
771017	.11	.076	.16	.038	.019	13
	.086	.066	.13	.033	.020	9
771118	.11	.077	.15	.054	.054	17
	.10	.076	.15	.053	.055	17

(Continued)

Table 10 (Continued)

Sample Date[i]	n-C_{15}	n-C_{16}	n-C_{17}	Pristane	Phytane	UCM[ii]
771212	.060	.027	.082	.033	.027	13
	.081	.038	.081	.038	.038	15
780220	.077	.038	.099	.13	.099	74
	.062	.041	.098	.12	.088	56
780428	.27	.17	.33	.17	.099	139
	.23	.15	.34	.16	.11	102
780523	.19	.90	.28	.081	.090	137
	.13	.58	.26	.091	.090	120
780630	.14	.081	.25	.059	.035	107
	.21	.098	.32	.075	.058	121
780721	.18	.11	.29	.048	N.D.	115
	.22	.15	.28	.025	N.D.	105
780828	.18	.079	.28	N.D.	N.D.	86
	.16	.11	.35	N.D.	N.D.	97
781020	.12	.045	.11	N.D.	N.D.	49
	.13	.046	.11	N.D.	N.D.	54
781121	.062	.027	.083	N.D.	N.D.	59
	.050	.014	.096	N.D.	N.D.	53
781228	.12	.054	.18	.060	N.D.	102
	.079	.055	.14	.046	N.D.	83

[i] See Table 1.

[ii] Not measured due to interference from internal standard or instrument recorder problem.

[iii] Not detected at $.005 \times 10^{-6}$ g/g dry weight.

[iv] UCM = Unresolved complex mixture signal in gas chromatogram; fl only.

TABLE 11

Selected Aromatic Hydrocarbon Concentrations in Narragansett Bay Mussels (Mytilus edulis) (10^{-9} g/g dry weight).

Compound	760609[i]	760720	760823	760909	761020	761111
Naphthalene	1	--[iii]	1	9	1	2
C_1 - Naphthalenes	3	2	7	10	--	7
C_2 - Naphthalenes	13	8	27	13	5	12
Phenanthrene	38	20	16	24	27	23
C_1 - Phenanthrenes	97	65	34	30	29	59
C_2 - Phenanthrenes	116	58	51	66	50	61
Fluoranthene	60	26	20	70	42	60
Pyrene	28	25	23	60	58	59
Chrysene[iv]	12	--	--	7	--	19
Benzopyrene[iv]	--[iii]	--	--	--	--	--
Dibenzothiophene	3	9	--	--	--	--
C_1 - Dibenzothiophenes	22	18	--	--	--	--
Total	393	232	180	288	212	304
% Recovery of Internal Standards[ii]						
Hexamethylbenzene	22	7	38	73	37	31
Hexaethylbenzene	--	--	--	--	--	--

(Continued)

Table 11 (Continued)

Compound	770623	770718	770810	770914	771017	771118	771212
Naphthalene	--	1	1	1	2	1	--
C_1 - Naphthalenes	--	5	1	2	5	7	9
C_2 - Naphthalenes	3	23	2	4	12	21	43
Phenanthrene	17	18	7	3	22	34	41
C_1 - Phenanthrenes	28	64	10	5	60	123	127
C_2 - Phenanthrenes	42	70	3	4	83	160	149
Fluoranthene	80	30	26	10	46	41	28
Pyrene	63	43	30	11	60	69	33
Chrysene[iv]	3	11	6	2	25	39	7
Benzopyrene[iv]	--	6	8	--	27	54	--
Dibenzothiophene	10	20	1	--	15	34	42
C_1 - Dibenzothiophenes	26	106	3	--	69	164	183
Total	271	396	97	42	426	748	662
% Recovery of Internal Standards							
Hexamethylbenzene	31	21	25	23	31	44	34
Hexaethylbenzene	87	43	55	48	50	58	40

(Continued)

Table 11 (Continued)

Compound	780220	780428	780523	780630	780721	780828
Naphthalene	--	2	1	2	--	--
C_1 - Naphthalenes	9	5	18	3	--	--
C_2 - Naphthalenes	35	22	68	13	--	--
Phenanthrene	63	52	47	23	27	3
C_1 - Phenanthrenes	185	47	142	48	41	--
C_2 - Phenanthrenes	330	104	200	67	37	--
Fluoranthene	127	51	47	48	95	14
Pyrene	81	32	22	32	47	19
Chrysene[iv]	112	--	--	--	--	--
Benzopyrene[iv]	104	--	--	--	--	--
Dibenzothiophene	58	--	80	14	--	--
C_1 - Dibenzothiophenes	198	--	388	60	--	--
Total	1300	316	1015	311	247	36.0
% Recovery of Internal Standards						
Hexamethylbenzene	32	38	41	27	33	14
Hexaethylbenzene	49	51	51	55	62	27

(Continued)

Table 11 (Continued)

Compound	781020	781121	781228
Naphthalene	--	4	--
C_1 -Naphthalenes	--	--	--
C_2 -Naphthalenes	--	--	--
Phenanthrene	9	6	6
C_1 - Phenanthrenes	--	8	15
C_2 - Phenanthrenes	--	4	14
Fluoranthene	30	18	38
Pyrene	25	16	30
Chrysene[iv]	--	--	37
Benzopyrene[iv]	--	--	33
Dibenzothiophene	--	--	
C_1 - Dibenzothiophenes	--	--	
Total	64	56	172
% Recovery of Internal Standards			
Hexamethylbenzene	11	15	11
Hexaethylbenzene	37	20	27

[i]See Table 1 for sample date code.

[ii]See discussion in methodology section.

[iii]Non-detected at 0.5×10^{-9} g/g dry weight.

[iv]Total of chrysene + aromatic hydrocarbons with m/e molecular ion of 228; total of benzopyrene + aromatic hydrocarbons with m/e molecular ion of 252.

TABLE 12

U.S. EPA Mussel Watch: West Coast PCB + DDE Data - 1976, 1977

	PCB 10^{-9} g/g Dry Weight		DDE 10^{-9} g/g Dry Weight	
	1976	1977	1976	1977
California				
San Diego Harbor (4 stations)	360-1400 Ave. 863	800	57	44
Point La Jolla	84	45	45	40
Oceanside Jetty	78	73	81	89
Santa Catalina Island	57	35	87	35
San Pedro Harbor* (5 stations)	440-8700 Ave. 2980	500	330-17000 Ave. 4045	340
Point Fermin	250	270	2500	1400
Rincon Cliffs	130	67	180	52
San Simeon	10	15	36	20
Point Arguello	39	26	67	71
Diablo Canyon	21	13	25	19
Soberanes	15	10	60	24
Santa Cruz	21	18	76	28
Farallon Islands	27	23	48	16
S. San Francisco bay	590	990	20	57
Point Arena	7	29	14	19
Cape Mendocino	10	4.6	2	3
Point St. George	3	8	8	4.8
Humbolt	17	7	4	3

(Cont.)

Table 12 (Continued)

	PCB		DDE	
	10^{-9} g/g Dry Weight		10^{-9} g/g Dry Weight	
	1976	1977	1976	1977
Oregon				
Gold Beach	24	10	8	4.7
Coos Bay	19	13	2	2.7
Yaquina Head	20	21	5	4.3
Tillamook Bay* (5 stations)	9-25 Ave. 16	25	2-17	8.6
Columbia River	42	26	2	5.2
Washington				
Willapa Bay* (2 stations)	21,43 Ave. 32	20	4,10	4.8
Grays Harbor	18	23	2	4.7
Cape Flattery	7	12	2	2.4
Puget Sound	41	37	2	3.3
Boundary Bay	36	41	3	5.0

*Stations composited in 1977; 1976 values are ranges and average.

TABLE 13

U.S. EPA Mussel Watch: East and Gulf Coast Data for Hydrocarbons, PCBs, and DDE in Mussels (Mytilus edulis) and Oysters (Crassostrea virginica, Ostrea equestrias)

Sample Location	Total of fl GC Peaks		fl UCM 10^{-6} g/g Dry Wt.		Aromatics 10^{-9} g/g Dry Wt.		PCB 10^{-9} g/g Dry Wt.		DDE 10^{-9} g/g Dry Wt.	
	1976	1977	1976	1977	1976	1977	1976	1977	1976	1977
Mussels										
Maine:										
Blue Hill Falls	.69	.83	< 5	< 5	45	--*	15	34	3.5	3.1
Sears Island	1.7	2.3	47	20	234	124	79	105	5.5	4.0
Cape Newagen	.59	1.8	9	< 5	48	--	55	85	4.6	4.8
Bailey Island	.68	.65	9	8	53	--	59	109	8.8	3.9
Portland	2.1	2.2	115	82	270	85	95	110	< 8	3.8
Massachusetts:										
Cape Ann	.44	1.4	< 3	< 5	30	--	96	112	7.4	4.2
Boston	6.7	8.7	298	311	3610	2100	635	735	< 44	< 26
Plymouth	1.8	1.4	18	17	66	--	226	366	< 16	< 17
Cape Cod Canal	.86	.52	< 3	7	41	--	215	188	< 11	< 7
Rhode Island:										
Sakonnet	.58	.66	< 3	< 5	32	--	120	177	< 11	< 7
Block Island	1.6	1.9	5	< 5	178	34	102	119	7.9	< 5
Connecticut:										
Millstone	.86	.90	37	20	110	--	321	184	< 16	< 8
New Haven	.36	1.2	21	15	163	56	129	299	< 7	< 9

(Cont.)

Table 13 (Continued)

Sample Location	Total of fl GC Peaks		fl UCM 10^{-6} g/g Dry Wt.		Aromatics 10^{-9} g/g Dry Wt.		PCB 10^{-9} g/g Dry Wt.		DDE 10^{-9} g/g Dry Wt.	
	1976	1977	1976	1977	1976	1977	1976	1977	1976	1977
Mussels										
New York:										
Manhasset Neck	.82	.83	39	25	620	193	838	700	< 30	< 31
Herod Point	.48	1.4	7	< 5	122	--	316	317	< 18	16
Fire Island	.77	.99	20	< 3	74	--	243	133	< 21	8
Great Gull Island	.53	--	13	--	73	--	272	--	< 14	--
Rockaway Point	1.7	2.4	82	101	307	137	575	476	< 32	< 20
New Jersey:										
Atlantic City	.39	2.3	5	< 5	134	--	348	244	21	12
Cape May	.35	73	19	6	208	144	334	259	24	26
Delaware:										
Cape Henlopen	.84	2.4	8	6	146	--	284	329	45	37
Maryland:										
Assateague	1.2	2.9	13	< 5	128	--	150	116	16	15
Virginia:										
Wachapreague Island	1.0	2.2	< 3	< 3	104	--	136	42	15	7.8

(Cont.)

Table 13 (Continued)

Sample Location	Total of fl GC Peaks		fl UCM 10^{-6} g/g Dry Wt.		Aromatics 10^{-9} g/g Dry Wt.		PCB 10^{-9} g/g Dry Wt.		DDE 10^{-9} g/g Dry Wt.	
	1976	1977	1976	1977	1976	1977	1976	1977	1976	1977
Oysters										
Virginia:										
Cape Charles	1.1	1.7	< 3	< 5	197	94	32	20	30	13
Lynnhaven Bay	1.2	.84	12	< 5	350	613	162	54	19	6.2
Wachapreague Island	.29	.83	< 3	< 5	30	--	84	77	13	9.7
North Carolina:										
Hatteras Island	.60	1.9	< 3	< 3	104	--	15	16	4.7	2.3
Beaufort	1.1	1.9	27	16	595	694	38	30	14	4.0
Cape Fear	.70	.68	< 3	< 3	66	--	30	18	7.2	4.0
South Carolina:										
Charleston	2.9	1.6	93	38	1720	400	82	62	15	< 4
Georgia:										
Sapelo Island	.51	.45	< 3	< 5	72	--	17	21	5.3	4.4
Savannah River	1.2	41	33	28	811	169	33	45	6.6	4.1

(Cont.)

Table 13 (Continued)

Sample Location	Total of fl GC Peaks		fl UCM 10^{-6} g/g Dry Wt.		Aromatics 10^{-9} g/g Dry Wt.		PCB 10^{-9} g/g Dry Wt.		DDE 10^{-9} g/g Dry Wt.	
	1976	1977	1976	1977	1976	1977	1976	1977	1976	1977
Oysters										
Florida:										
St. Augustine	1.6	.74	5	< 5	268	34	149	91	< 10	< 4
Naples	.31	1.4	< 3	< 5	31	--	21	16	6.0	2.8
Ancelote	1.6	1.5	< 3	< 5	190	--	77	53	9.5	5.6
Cedar Key	.45	1.2	< 3	< 5	68	--	< 20	27	5.5	3.2
Apalachicola	.70	.32	< 3	< 5	78	--	< 20	48	15	7.0
Panama City	2.2	2.1	< 3	-15	188	--	< 20	131	28	28
Pensacola	.67	3.1	8	< 5	221	--	107	46	23	10
Alabama:										
Dauphin Island		.87	< 3	< 3	161	25	< 20	94	42	24
Oysters										
Mississippi:										
Pass Christian	.40	2.2	5	5	167	--	< 20	35	11	5.7
Biloxi	2.5	4.3	79	209	610	--	336	297	28	< 13

*Not analyzed.

TABLE 14

U.S. EPA Mussel Watch
Three Year Data Set PCB Concentrations

Year →	PCB 10^{-9} g/g Dry Weight 1976	1977	1978
Station Location			
Sears Island, Maine	79	105	69
Bailey Island, Maine	59	109	41
Cape Ann, Massachusetts	96	112	93
Plymouth, Massachusetts	226	366	328
Block Island, Rhode Island	102	119	72
New Haven, Connecticut	129	299	386
Rockaway, New York	575	476	303
Beaufort, North Carolina	38	30	< 15
Charleston, South Carolina	82	62	< 15
Pass Christian, Mississippi	< 20	35	< 15
Point La Jolla, California	84	45	77
San Pedro Harbor, California*	440 to 8700	500	328
Columbia River, Oregon	42	26	22
Boundary Bay, Washington	36	41	16

*5 Stations composited in 1977, 1978; range for 1976.

Table 15

Concentrations of PCB's in Mussels - Mytilus edulis - from Massachusetts Coastal Area[i]

(10^{-9} g/g dry weight)

Sampling Location & Date	PCB Concentration As Arochlor 1016 or 1242	PCB Concentration As Arochlor 1254
New Bedford Harbor		
781002[iii]	16 x 10^3	15 x 10^3
Vineyard Sound		
Near Gayhead		
781011	--ii	95
781026	--	96
790828	--	98
790926	--	138
Nantucket Harbor		
(West Jetty)		
800717	--	80
800924	--	22
(Esther Island)		
800717	--	72
Cuttyhunk Island		
(West Pond)		
800628	--	176
800828	--	232
Cape Cod Canal		
760925		215
770819	--	188
800114	--	240
800214	--	258
800310	--	176
800424	--	200
800522	--	226

(Continued)

Table 15 (Continued)

Sampling Location & Date	PCB Concentration	
	As Arochlor 1016 or 1242	As Arochlor 1254
Plymouth		
760923	--	226
770818	--	366
780926	--	328
Boston		
760922	--	635
770818	--	735
Cape Ann		
760921	--	96
770817	--	112
780924	--	93

i See Figure 6.
ii None detected by packed column G.C. Predominantly Arochlor 1254.
iii Year, Month, Day.

TABLE 16

Hydrocarbon Data for 1976, 1977 West Coast and Louisiana-Texas Gulf Coast Samples*

Sample Location	Total Hexane Eluate GC Peaks (10^{-6} g/g Dry Weight) 1976	1977	Unresolved Complex Mixture 1976	1977
California				
San Diego Harbor (4 stations)	.32-62	60	++	+
Point La Jolla	18	5.7	+	+
Oceanside Jetty	7.6	8.1		
Santa Catalina Island	N.A.	8.1		++
San Pedro Harbor (5 stations)	.19-11	70	++	+
Point Fermin	4.9	11		++
Rincon Cliffs	8.6	28	-	+
San Simeon	N.A.	43		+
Point Arguello	N.A.	12		+
Diablo Canyon	.31	68		-
Soberanes	1.3	107		-
Santa Cruz	3.6	6.5	+	+
Farallon Islands	11	3.8		-
S. San Francisco Bay	.35-4.4	14		++
N. San Francisco bay	N.A.			
Point Arena	.66	4.6		++
Cape Mendocino	.29	21		-
Point St. George	4.1	8.2	+	-
Humbolt	1.3	7.2		-
Oregon				
Gold Beach	.96	3.9		+
Coos Bay	15	1.9		+
Yaquina Head	9.3	22		+
Tillamook Bay (5 stations)	.59-96	13	+	++
Columbia River	1.9	30		+
Washington				
Willapa Bay	1.4	44		-
Grays Harbor	3.9	27		-
Cape Flattery	8.6	3.2		-
Puget Sound	N.A.	20		++
Boundary Bay	.36	3.6	+	-

(Continued)

Table 16 (Continued)

Sample Location	Total Hexane Eluate GC Peaks (10^{-6} g/g Dry Weight) 1976	1977	Unresolved Complex Mixture 1976	1977
Louisiana				
Lake Borgne	.87	x		
Drum Bay	x	36		++
Lake Campo	1.1	x	++	
Pumpkin Bay	x	3.9		++
Black Bay	2.6	30		++
California Bay	x	x		
Quarantine Bay	x	8.0		++
Bastian Bay	1.0	x	++	
Barataria Bay	1.4	x		
Bay de Islettes	1.1	x		
Golden Meadows		5.6		++
Lake Barre	.40	x		
Terrabonne Bay	x	x		
Bay Cheland	x	x		
Junop Bay	x	17		++
Bayou de West	2.5	x		
Calcasieu Lake	4.5	17		++
Texas				
Lake Sabine	1.8	16		++
Galveston Bay	.54	3.9	++	++
East Matagordo Bay	1.6	9.0		+
Matagordo Bay	2.2	15	++	+
Lavaca Bay, North	x	17		++
Lavaca Bay	x	6.4		++
San Antonio Bay	.97	8.6		-
Panther Point	.34	x		
Espiritu Santo Bay	.96	x	+	
Mesquite Bay	x	5.5		+
Fulton	1.4	x		
Aransas Bay	x	37		+
Brownsville	x	4		++

*Empty columns - no data available for the parameter for those samples.

x = Sample not collected that year.

- = UCM not significant.

+ = UCM significant.

++ = UCM major part of GC.

TABLE 17

Three Year Data
U.S. EPA Mussel Watch

Year →	fl UCM Hydrocarbons 10^{-6} g/g Dry Weight		
	1976	1977	1978
Sears Island, Maine	47	20	32
Bailey Island, Maine	9	8	18
Cape Ann, Massachusetts	< 3	< 5	20
Plymouth, Massachusetts	18	17	30
Block Island, Rhode Island	5	< 5	10
New Haven, Connecticut	21	15	44
Rockaway, New York	82	101	72
Beaufort, North Carolina	27	16	84
Charleston, South Carolina	93	38	46
Pass Christian, Mississippi	5.0	5	8

TABLE 18a

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	BHF 760915	SI 760916	CN 760917	BI 760918	PORT 760920	CA 760921
Naphthalene	8	3	--	--	3	--
C_1 - Naphthalene	9	2	1	--	9	--
C_2 - Naphthalene	5	6	3	--	3	--
Phenanthrene	8	30	10	6	31	2
C_1 - Phenanthrene	6	33	10	12	34	3
C_2 - Phenanthrene	--	61	10	15	71	6
Fluoranthene	5	47	8	12	64	12
Pyrene	3	51	5	8	56	6
Chrysene[i]	--	--	--	--	--	--
Benzopyrene[i]	--	--	--	--	--	--
Dibenzothiophene	--	--	--	--	--	--
C_1 - Dibenzothiophene	--	--	--	--	--	--
Total	45	234	48	53	270	31
% Recovery of Internal Standards						
Hexamethylbenzene	79	48	22	4	42	0
Hexaethylbenzene	--	--	--	--	--	--

TABLE 18b

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	B 760922	PLY 760923	CCC 760925	SK 760927	BLK 761001	GG 761011
Naphthalene	4	1	2	--	1	1
C_1 - Naphthalene	29	2	--	--	--	2
C_2 - Naphthalene	204	2	--	--	23	--
Phenanthrene	91	16	14	10	20	10
C_1 - Phenanthrene	587	10	--	6	33	8
C_2 - Phenanthrene	1820	5	2	--	41	12
Fluoranthene	240	16	12	12	37	18
Pyrene	329	14	12	4	23	16
Chrysene[i]	13	--	--	--	--	6
Benzopyrene[i]	--	--	--	--	--	--
Dibenzothiophene	90	--	--	--	--	--
C_1 - Dibenzothiophene	200	--	--	--	--	--
Total	3610	66	41	32	178	73
% Recovery of Internal Standards						
Hexamethylbenzene	68	14	30	14	31	15
Hexaethylbenzene	--	--	--	--	--	47

TABLE 18c

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	MIL 761004	NH 761006	HP 761010	MN 761009	RK 761012	FI 761011
Naphthalene	1	4	1	--	3	2
C_1 - Naphthalene	--	5	2	--	3	8
C_2 - Naphthalene	--	4	4	--	8	3
Phenanthrene	18	20	15	18	24	13
C_1 - Phenanthrene	15	14	18	23	32	11
C_2 - Phenanthrene	16	27	24	84	56	--
Fluoranthene	32	34	34	114	67	20
Pyrene	28	48	21	381	107	12
Chrysene[i]	--	8	3	--	9	3
Benzopyrene[i]	--	--	--	--	--	2
Dibenzothiophene	--	--	--	--	--	--
C_1 - Dibenzothiophene	--	--	--	--	--	--
Total	110	163	122	620	307	74
% Recovery of Internal Standards						
Hexamethylbenzene	17	31	15	38	24	45
Hexaethylbenzene	42	--	38	--	--	--

TABLE 18d

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	AC 761004	MAY 761015	CH 761104	A 761106	W 761116 mu	W 761116 oy
Naphthalene	2	2	--	1	8	2
C_1-Naphthalene	3	1	--	3	6	3
C_2-Naphthalene	4	2	--	3	--	--
Phenanthrene	16	26	23	17	28	9
C_1-Phenanthrene	14	28	21	21	19	9
C_2-Phenanthrene	27	42	37	36	8	--
Fluoranthene	35	50	32	27	23	5
Pyrene	31	50	30	18	12	4
Chrysene[i]	3	6	3	3	--	--
Benzopyrene[i]	--	--	--	--	--	--
Dibenzothiophene	--	--	--	--	--	--
C_1-Dibenzothiophene	--	--	--	--	--	--
Total	134	208	146	128	104	30
% Recovery of Internal Standards						
Hexamethylbenzene	17	19	0	19	30	41
Hexaethylbenzene	45	--	--	--	--	--

TABLE 18e

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	CC 761109	LH 761115	H 761119	BU 761121	FR 761122	CHR 761201
Naphthalene	3	2	0	1	2	5
C_1 - Naphthalene	3	6	1	4	4	12
C_2 - Naphthalene	7	18	4	8	4	48
Phenanthrene	29	47	6	63	15	85
C_1 - Phenanthrene	31	41	9	78	12	258
C_2 - Phenanthrene	54	52	18	142	9	616
Fluoranthene	47	106	42	169	13	258
Pyrene	19	62	23	118	8	251
Chrysene[i]	5	4	2	11	--	32
Benzopyrene[i]	--	--	--	--	--	--
Dibenzothiophene	--	6	--	--	--	20
C_1 - Dibenzothiophene	--	7	--	--	--	133
Total	197	350	104	595	66	1720
% Recovery of Internal Standards						
Hexamethylbenzene	30	15	1	15	32	24
Hexaethylbenzene	51	38	--	--	38	--

TABLE 18f

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	SAV 761130	SAP 761129	STA 761204	NAP 761223	AN 761230	KEY 761231
Naphthalene	4	5	2	1	1	--
C_1 - Naphthalene	6	6	6	1	4	2
C_2 - Naphthalene	28	6	12	3	18	5
Phenanthrene	92	15	31	3	14	11
C_1 - Phenanthrene	86	13	46	5	24	9
C_2 - Phenanthrene	192	5	62	4	60	9
Fluoranthene	152	16	62	8	33	20
Pyrene	212	7	39	7	33	10
Chrysene[i]	7	--	8	--	3	2
Benzopyrene[i]	--	--	--	--	--	--
Dibenzothiophene	10	6	--	--	--	--
C_1 - Dibenzothiophene	18	--	--	--	--	--
Total	811	72	268	31	190	68
% Recovery of Internal Standards						
Hexamethylbenzene	30	29	55	2	16	20
Hexaethylbenzene	--	37	--	--	37	42

TABLE 18g

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	AP 770104	PAN 770105	PEN 770108	DAU 770113	BIL 770117	PASS 770115
Naphthalene	1	2	1	1	6	--
C_1 - Naphthalene	3	3	4	3	14	--
C_2 - Naphthalene	4	9	7	8	65	10
Phenanthrene	18	36	5	22	43	34
C_1 - Phenanthrene	19	38	42	34	120	26
C_2 - Phenanthrene	17	44	44	64	258	50
Fluoranthene	10	36	70	18	42	26
Pyrene	6	17	40	11	63	18
Chrysene[i]	--	2	9	--	--	2
Benzopyrene[i]	--	--	--	--	--	--
Dibenzothiophene	--	--	--	--	--	--
C_1 - Dibenzothiophene	--	--	--	--	--	--
Total	79	188	221	161	610	167
% Recovery of Internal Standards						
Hexamethylbenzene	20	32	22	14	35	32
Hexaethylbenzene	--	60	--	44	--	--

TABLE 18b

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	SI 770813	P 770816	B 770818	BLK 770825	NH 770827	MN 770828	RK 770830
Naphthalene	1	--	15	2	1	--	1
C_1-Naphthalene	4	--	59	6	3	--	5
C_2-Naphthalene	8	--	109	4	3	--	2
Phenanthrene	10	10	310	6	7	7	15
C_1-Phenanthrene	15	15	247	--	5	11	22
C_2-Phenanthrene	18	10	552	--	--	11	31
Fluoranthene	35	26	336	9	14	49	30
Pyrene	25	19	248	8	15	78	22
Chrysene[i]	8	7	92	--	7	28	8
Benzopyrene[i]	--	--	50	--	--	11	--
Dibenzothiophene	--	--	24	--	--	--	--
C_1-Dibenzothiophene	--	--	61	--	--	--	--
Total	124	85	2100	34	56	193	137
% Recovery of Internal Standards							
Hexamethylbenzene	33	23	31	41	17	14	36
Hexaethylbenzene	57	34	28	63	17	86	60

TABLE 181

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	MAY 770902	LH 770910	CC 770911	BU 770914	CHR 770918	SAV 770920	ST.A 770923	DAU 771008
Naphthalene	4	--	1	1	1	1	2	1
C_1 -Naphthalene	7	--	7	2	6	4	2	1
C_2 - Naphthalene	8	3	6	3	7	7	2	1
Phenanthrene	14	56	12	93	22	14	4	5
C_1 - Phenanthrene	11	29	10	53	18	30	4	+
C_2 - Phenanthrene	6	35	5	45	38	29	2	2
Fluoranthene	48	234	28	235	127	22	10	3
Pyrene	36	156	16	165	132	54	6	3
Chrysene[i]	11	74	9	82	41	6	3	--
Benzopyrene[i]	--	19	--	16	10	--	--	--
Dibenzothiophene	--	4	--	--	--	--	--	1
C_1 - Dibenzothiophene	--	4	--	--	--	--	--	6
Total	144	613	94	694	400	169	34	25
% Recovery of Internal Standards								
Hexamethylbenzene	58	18	20	39	43	57	46	31
Hexaethylbenzene	92	47	37	46	57	88	59	66

TABLE 18j

Selected Aromatic Hydrocarbon Concentrations in Mytilus edulis and Crassostrea virginica from the U.S. East and Gulf Coasts, 1976, 1977, 1978
(10^{-9} g/g dry weight)

Compound	SPH 780428	NB 781002	RK 781015	BEAU 781029	CHR 781030	ARS 781121
Naphthalene	1	2	7	2	--	--
C_1 - Naphthalene	2	--	5	8	--	--
C_2 - Naphthalene	3	--	4	31	--	10
Phenanthrene/Anthracene	23/39	10	46	72/16	15	26
C_1 - Phenanthrene	95	21[ii]	55	92	10	131
C_2 - Phenanthrene	107	--	41	118	--	316
Fluoranthene	112	124	44	206	97	11
Pyrene	58	71	42	104	74	11
Chrysene[i]	219	79	88	144	78	--
Benzopyrene[i]	233	129[ii]	51	78	45	--
Dibenzothiophene	--	--	--	--	--	--
C_1 - Dibenzothiophene	--	--	--	--	--	--
Total	892	436[ii]	383	870	319	504
% Recovery of Internal Standards						
Hexamethylbenzene	23	30	51	43	31	23
Hexaethylbenzene	30	37	67	41	41	50

[i] See Table II, footnote iv.
[ii] Concentration may be high due to interference from PCBs co-eluting.

TABLE 19

Three Year Data for Aromatic Hydrocarbons
(10^{-9} g/g dry weight)

	Rockaway Point, N.Y.			Beaufort, N.C.			Charleston, S.C.		
	1976	1977	1978	1976	1977	1978	1976	1977	1978
Naphthalene	3	1	7	1	1	2	5	1	--
C_1 - Naphthalenes	3	5	5	4	2	8	12	6	--
C_2 - Naphthalenes	8	2	4	8	3	31	48	7	--
Phenanthrene	24	15	46	63	93	72	85	22	15
C_1 - Phenanthrenes	32	22	55	78	53	92	258	18	10
C_2 - Phenanthrenes	56	31	41	142	45	118	616	38	--
Fluoranthene	67	30	44	169	235	206	258	127	97
Pyrene	107	22	42	118	165	104	251	132	74